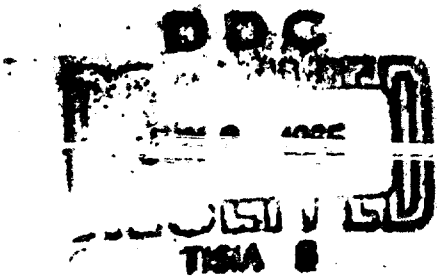
FINAL REPORT

"Fabrication and Properties of Tungsten Single Crystals"



Part I

"Growth of Single Crystal Tungsten To Be Used for Rolling Into Single Crystal Sheet"

Work Done by: G. W. Edwards, J. W. Arthur

Work Reported by: G. W. Edwards

UNION CARBIDE CORPORATION, Linde Division - Prime Contractor

Part II

"Rolled Single Crystal Tungsten Sheet"

Prepared by: L. Raymond

AERONUTRONIC DIVISION, Philco Corporation - Subcontractor

Prepared under Contract NOw 64-0055-c
For the Materials Branch,
Bureau of Naval Weapons
Department of the Navy

For period March 1, 1964 to February 26, 1965

May 14, 1965

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SUMMARY

This report is the Final Technical Report of work performed under Bureau of Naval Weapons Contract NOw 64-0055-c by Linde Division of Union Carbide Corporation and its subcontractor Aeronutronic Division of Philco Corporation. The work is concerned with the deformation of tungsten single crystals.

The objective of this study has been to find a method for deforming single crystals of tungsten such that monocrystallinity is maintained throughout the deformation process. This objective has been met. The technology of deformation and joining of tungsten single crystals has now reached the point, through this study, where simple items of hardware which retain their single crystal character can be fabricated.

An item of hardware which is also a single crystal is desirable in certain applications for two reasons. The recrystallization phenomenon with its associated brittleness is absent. Thus, the single crystal will remain ductile even if heated to the melting point. Because of the single crystallinity of material, it can be welded by a state-of-the-art technique and maintain its single crystallinity even in the weld bead. Thus, recrystallization and brittleness associated with weldments are also avoided.

The method for cold deformation of tungsten single crystals requires extreme care in its present state of development. The starting single crystal boules from which billets are fabricated must be of high purity and of low dislocation concentration and lineage. In order to maintain monocrystallinity, single or duplex slip must be employed. Therefore, careful orientation of the starting billet is necessary in order that the desired slip system alone be active. For deformation by rolling, the billet orientations (110)[001] or (100)[011] are necessary to encourage duplex slip. Other orientations allow multiple slip and do not yield single crystals. Furthermore, deformation of less than 10% per pass is necessary. If deformations of more than 10% are produced, multiple slip will occur even on properly oriented billets. Periodically, the surface of the billet must be chemically polished so that multiple slip caused by friction between the billet surface and the rolls is removed. If this surface is not removed, then subsequent recovery heat treatment will cause recrystallization of this highly strained area.

Many high temperature applications for metals could conceivably be filled by fabricated tungsten single crystals. For example, a rocket nozzle could be constructed from rolled single crystal sheets which are assembled and welded into the proper configuration. Other operations such as press forging and shear

spinning, if useable, could produce turbine buckets and rocket nose cones in single crystal form.

The strength of tungsten single crystals could be increased in order to counteract its high density. Such means as solid solution hardening or precipitation hardening mechanisms could be invoked which would allow tungsten single crystals to be of even more practical use in high temperature applications.

Thus, because of the successful deformation and welding of tungsten single crystals without the loss of monocrystallinity, a new area of possible applications for tungsten is open. This work should be continued in order to rapidly exploit the advances made so far.

I. SUMMARY OF CRYSTAL GROWTH:

This is the final report describing the work done for the Materials Branch, Bureau of Naval Weapons under Contract NOw 64-0055-c from March 1, 1964 to February 26, 1965.

Seventy tungsten and seven tungsten-0.6% niobium crystals were grown by the Arc-Verneuil method using high purity commercial tungsten powder. From these, sixteen pure tungsten and two niobium bearing crystals were selected for machining into rectangular billets. These billets were believed, because of their low crystal misorientation and deviation of crystal orientation from cylindrical axis, to be the most likely to survive cold rolling. The Arc-Verneuil technique was also used to grow three slab-shaped tungsten crystals 3/8-inch x 1-1/2-inches x 4-inches for evaluation pertaining to crystal perfection.

II. INTRODUCTION:

Experimental data obtained from the previous investigations showed that it was possible to roll single crystal tungsten and still retain its monocrystallinity.[1] It was also shown that certain crystallographic orientations, specifically the (001)[110] and (110)]100] orientations, possess superior qualities to others for rolling. Thus, it was deemed possible to obtain single crystal tungsten sheet from properly oriented crystals by appropriate roll deformation, anneal cycles. Since it is known that metallic impurities tend to raise the recrystallization temperature, W-0.6% Nb single crystals of the same favorable orientation also will be subjected to the same conditions as the pure tungsten crystals for comparison studies.

In the interest of obtaining a wider sheet for better utility, a brief investigation was made concerning the feasibility of growing tungsten slabs of the high crystal quality necessary for rolling.

The portion of work under the contract performed by the Linde Division of Union Carbide Corporation at Speedway, Indiana, consisted of growing single crystals of tungsten or its alloys, evaluating the grown crystals as to their perfection and orientation, and fabricating them into billets suitable for rolling. Aeronutronic Division of Philco Corporation performed the rolling of the billets and the evaluation of the rolled product. An account of this work is given in Part II of this report.

III. MASTER SEED PREPARATION

As was noted in earlier work, in order to grow high quality tungsten single crystals via the Arc-Verneuil method, it is imperative that seed stock be of the highest obtainable crystal quality and as close to the desired crystal direction as possible. Two crystals of high perfection ([110] and [001] orientations) to be used for seed stock were grown from seeds used in the previous program. These crystals were ground to smooth cylinders having one end surface normal to the crystal axis. This end was then electrolytically polished in a 5% NaOH solution to remove distorted material caused by the machining and grinding. A Laue Back-Reflection pattern was obtained and corrections were made to bring the cylinder axis and the desired growth direction in coincidence. Necessary machining techniques were then used to produce a sufficient length of the desired orientation. The seeds were then repolished and checked by the Laue technique to ascertain final seed orientation.

IV. CRYSTAL GROWTH

A. Tungsten Single Crystal:

The seed crystals prepared above, complemented by the seed stock from the previous program, were used to grow tungsten crystals via the Arc-Verneuil technique using commercially available high purity tungsten powder. Fifty-eight [110] and twelve [001] crystals were grown by this technique from which 8 each were selected for machining into billets on the basis of minimum crystal misorientation and deviation of crystal orientation from the cylindrical axis. The nominal size of the as-grown crystals was 7/16-inch diameter by 8-inches long, of which 6-1/2-inches was machined into billets and 1-1/2-inches was used for chemical analysis.

B. Tungsten-0.6% Niobium Single Crystals:

Seven tungsten-0.6% niobium crystals, [110] orientation, were grown by the Arc-Verneuil method from which two were selected for machining into billets. These crystals were grown with powder obtained by blending high purity tungsten and niobium powder in a stainless steel twin cone blender for four hours.

C. Slab Tungsten Crystals:

The slab-shaped tungsten crystals shown in Figure 1 were also grown by the Arc-Verneuil technique. The rectangular shape was obtained by reciprocating the arc source across the growth interface rather than holding it stationary as was done to grow the cylindrical crystals. The outward general

appearance of these slabs was good and after an electrolytic etch, only three or four lineage lines were visible. After machining a representative one, however, a larger number of lineage bands became visible and porosity was evident along the growth layers. For these reasons, this slab billet was not sent to Aeronutronic for rolling.

V. BILLET PREPARATION

A. Orientation and Fabrication:

On the crystals selected for rolling, a small flat 1/4-inch by 3/8-inch was ground on one end to be used as a reference surface when the Laue patterns were taken and to facilitate the mounting of the crystals on machining blocks. After results of the Laue shots were obtained, the crystal end with the flat was placed in an omni-vise and the necessary rotation around the cylindrical axis was made to bring the plane of desired crystal orientation to the horizontal position as viewed along the axis of rotation. The crystal was then mounted on standard machining blocks with DeKhotinsky cement. The crystal and block were then placed on the magnetic chuck of a surface grinder and ground along the desired rolling plane until a 3/8-inch wide flat was obtained along the length of the crystal. The crystal was demounted, rotated 180° and mounted again on the block for grinding of the opposite side to yield a crystal with parallel flats. The crystal could then be clamped in a parallel jaw vise for the remainder of the grinding operation. The dimensions of the machined billet were 1/4-inch x 3/8-inch x 6-1/2-inches. The billets fabricated in this manner are shown in Table I.

B. Surface Preparation:

The machined crystal billets prepared by the above method were chemically polished in 70% HF - 30% HNO_3 solvent to remove 0.005-inch to 0.008-inch material from each billet face which had been distorted due to grinding. It was felt that these surfaces, if not removed, would interfere in the rolling and recrystallization studies by contributing nucleating sources for premature recrystallization.

VI. CHEMICAL ANALYSIS OF CRYSTALS

The 1-1/2-inch length cut from the cap end of the crystal was carefully cleaved and the pieces obtained from the interior portion were used for chemical analysis. Carbon and oxygen were determined on the larger chips using a Leco conductometric analyzer. The smaller pieces were crushed in a Platner Diamond motor between platinum sheets and used for the determination of metallic impurities spectrographically using a 1.5 meter A.R.L. spectrograph and the Harvey-Semiquantitative method.[2] No analysis was made for nitrogen or hydrogen since

past results showed the content of each to be less than 1 ppm. The niobium in the W-0.6% Nb crystals was separated by ion-exchange techniques and determined gravimetrically. Analytical results are shown in Table II.

VII. ORIENTATION ANALYSIS BEFORE ROLLING

After the 6-1/2-inch crystal had been machined into the finished billet, a 1/2-inch sample was cut from the growth end and electrolytically polished and etched in a 5% sodium hydroxide solution. A Laue Pattern was taken to determine the deviation of the longitudinal axis of the crystal billet from the desired rolling direction and deviation of the billet rolling plane from the desired rolling plane. A Schulz-Wei photograph [3] was taken of each sample to determine the number and magnitude of low-angle grain boundaries present in the final billet. The low-angle grain boundaries observed in the billets are shown by two representative Schulz-Wei photographs, Figures 2 and 3. The results of the X-ray examination of the billet orientation and crystal structure imperfections are shown in Table III.

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2. C. E. Harvey, A Method of Semi-Quantitative Spectrochemical Analysis, Applied Research Laboratories, Glendale, California, 1948.

3. C. T. Wei, "Zone Reflection Camera for Studying Crystal Imperfections by the Schulz Technique", The Review of Scientific Instruments 27 (6), 397-399 (June 1956).

TABLE I

TUNGSTEN CRYSTALS SELECTED FOR ROLLING

Crystal Number	Rolling Plane	Rolling Direction
1690-22	(110)	[001]
1690-24	(110)	[001]
1690-27	(001)	[110]
1690-32	(001)	[110]
1690-34	(110)	[001]
1690-38	(110)	[001]
1690-39	(110)	[001]
1690-47	(001)	[110]
1690-50	(100)	[001]
1690-52	(100)	[001]
1690-53	(110)	[001]
1690-65	(001)	[110]
1690-71	(001)	[110]
1690-84	(001)	[110]
1690-88	(001)	[110]
1690-89	(001)	[110]
TUNGSTEN - 0.6% NIOBIUM		
1690-92	(001)	[110]
1690-96	(001)	[110]

TABLE II

CHEMICAL ANALYSIS OF TUNGSTEN CRYSTAL BILLETS

Crystal Number	C PPM	O PPM	WEIGHT PER CENT Al	Co	Cr	Cu	Fe	Mg	Mn	Mo	Ni	Sn	Nb	Si
1690-22	9	10	← Not Detected →											
1690-24	7	12	← Not Detected →											
1690-27	3	9	← Not Detected →											
1690-32	4	3												
1690-34	9	9						10^{-3}						
1690-38	3	12	← Not Detected →											
1690-39	3	10					10^{-3}	10^{-3}				10^{-2}		
1690-47	4	3	← Not Detected →											
1690-50	4	3						10^{-4}					10^{-2}	
1690-52	3	4	← Not Detected →											
1690-53	4	6					10^{-3}	10^{-4}					10^{-2}	
1690-65	4	2					10^{-3}	10^{-4}						10^{-3}
1690-71	5	3					10^{-3}							10^{-3}
1690-84	5	4					10^{-3}							
1690-88	6	2					10^{-2}							
1690-89	5	5	← Not Detected →											
1690-92*	6	8					10^{-3}						0.47%	
1690-96*	4	9	← Not Detected →										0.52%	
Nominal Lower Limits of Detectability			10^{-4}	10^{-3}	10^{-3}	10^{-4}	10^{-3}	10^{-4}	10^{-3}	10^{-2}	10^{-3}	10^{-3}	10^{-3}	10^{-3}

*Tungsten-0.6% Niobium Billets

Note: Metallic analysis semi-quantitative accuracy: $10^{-n} \Longrightarrow 10^{1-n}\% \leq C \leq 10^{-n}\%$

TABLE III

X-RAY ANALYSIS OF TUNGSTEN CRYSTAL BILLETS

Crystal Number	Deviation of Crystal Axis from Rolling Direction	Deviation of Rolling Plane Normal to Ground Flat	Maximum Low-Angle Grain Boundary
1690-22	1° ← 6° ↓	7°	5°
1690-24	3° → 0°	3°	50'
1690-27	5° → 7° ↓	2°	4°
1690-32	3° ← 1° ↑	0°	45'
1690-34	0° 2-1/2° ↓	2°	50'
1690-38	3° ← 3° ↓	5°	30'
1690-39	1° → 4° ↓	4°	2°
1690-47	5° → 7° ↑	3°	15'
1690-50	1° → 1° ↓	1°	1-1/2°
1690-52	1° ← 1-1/2° ↓	1-1/2°	1-1/2°
1690-53	0° 3° ↓	4°	1°
1690-65	5° ← 5° ↑	0°	1°
1690-71	3° → 4° ↑	1°	30' Strained Maximum
1690-84	3° ← 4° ↓	1°	30' " "
1690-88	7° ← 6° ↑	1°	30' " "
1690-89	5° ← 5° ↑	2°	30' " "
1690-92	5° → 8° ↑	2°	1°
1690-96	5-1/2° → 8° ↑	1°	25'

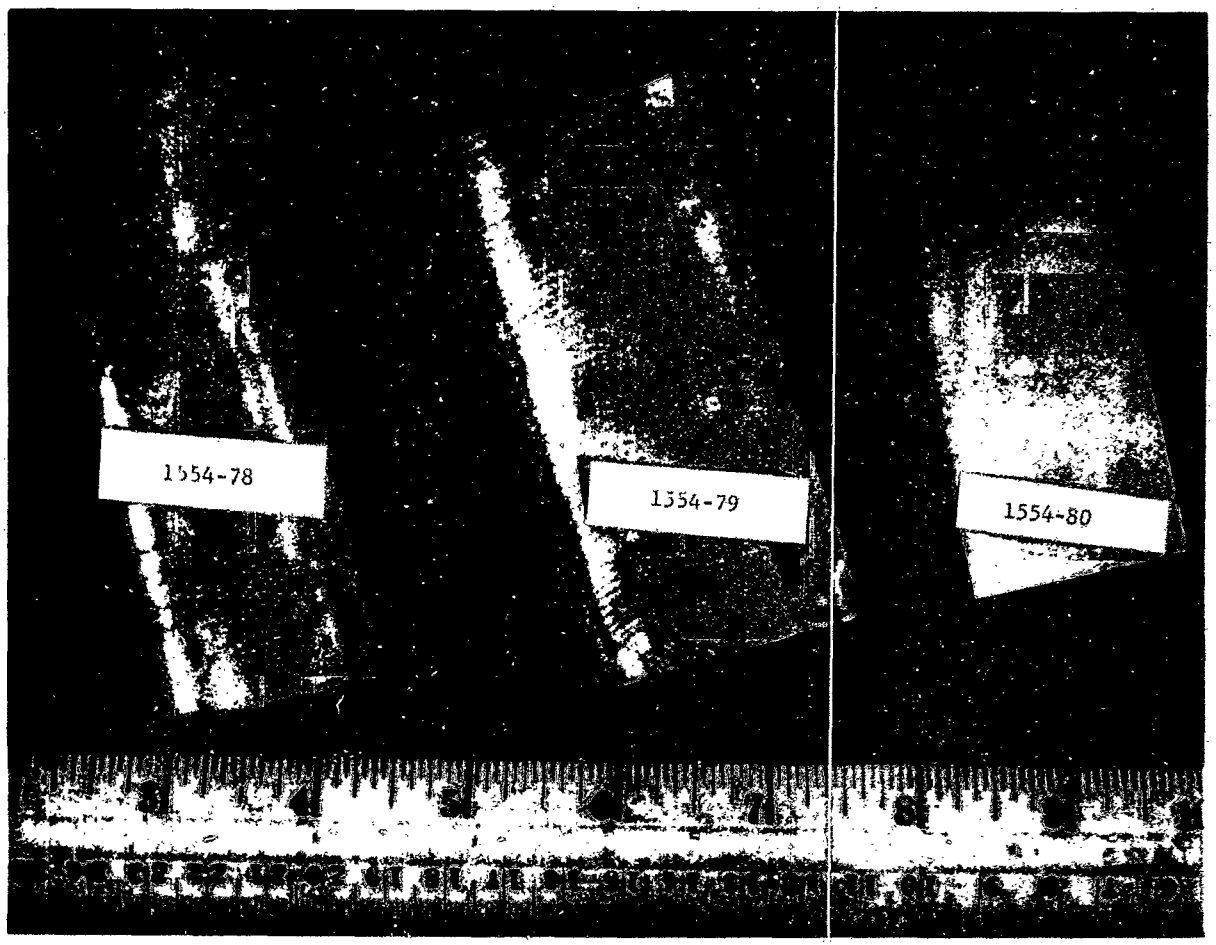

Figure 1. As-grown Tungsten Crystal Slab and Billet Machined from Slab.

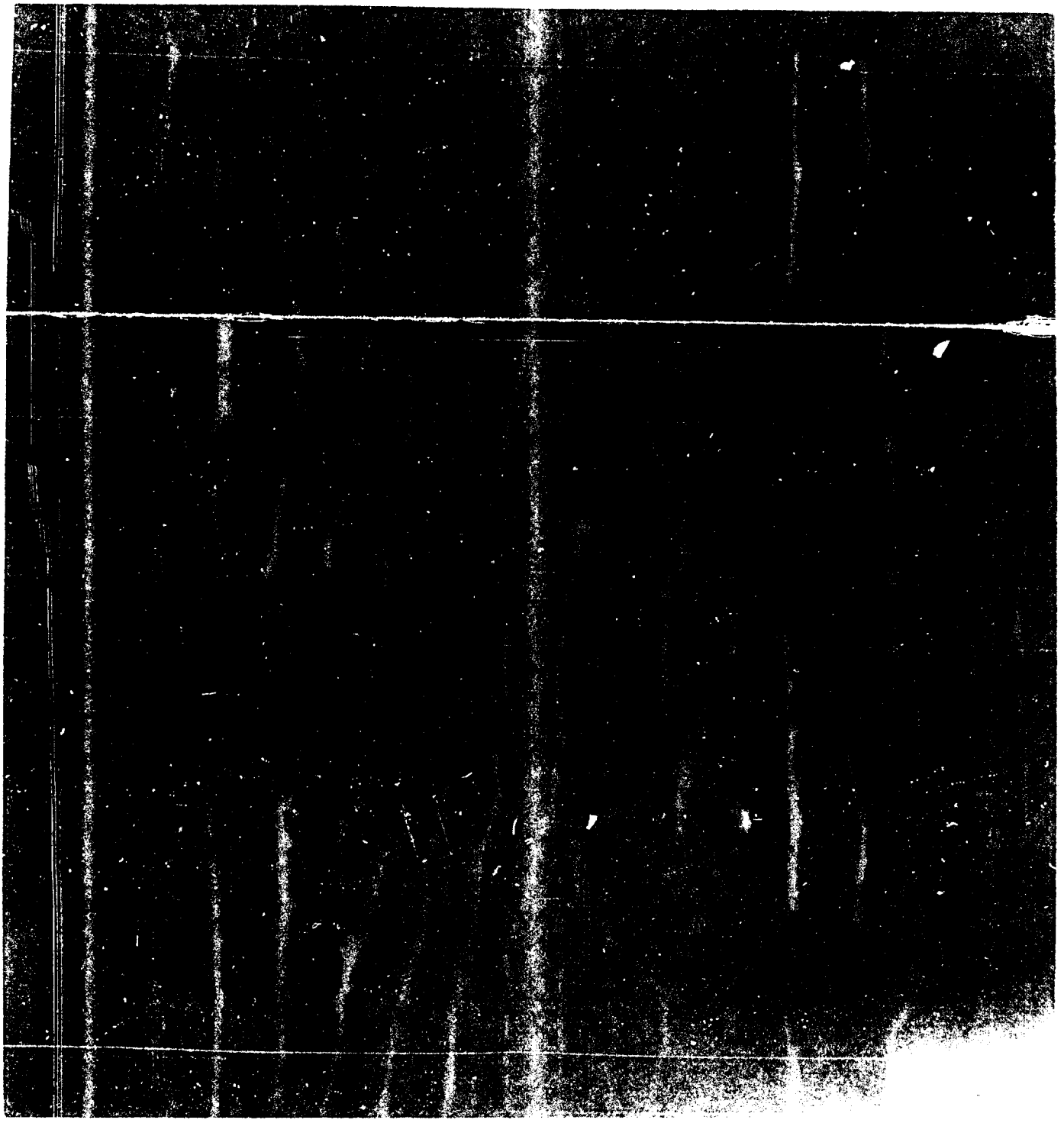

Figure 2. Schulz-Wei photo of 1690-38 (110)[001] cross-section. [001] is perpendicular to page, (110) normal lies along vertical in page. Bracket shows misorientation $< 1/2°$. There is larger misorientation of $\sim 1°$ at corner of billet. White lines on image are characteristic radiation from tungsten target.

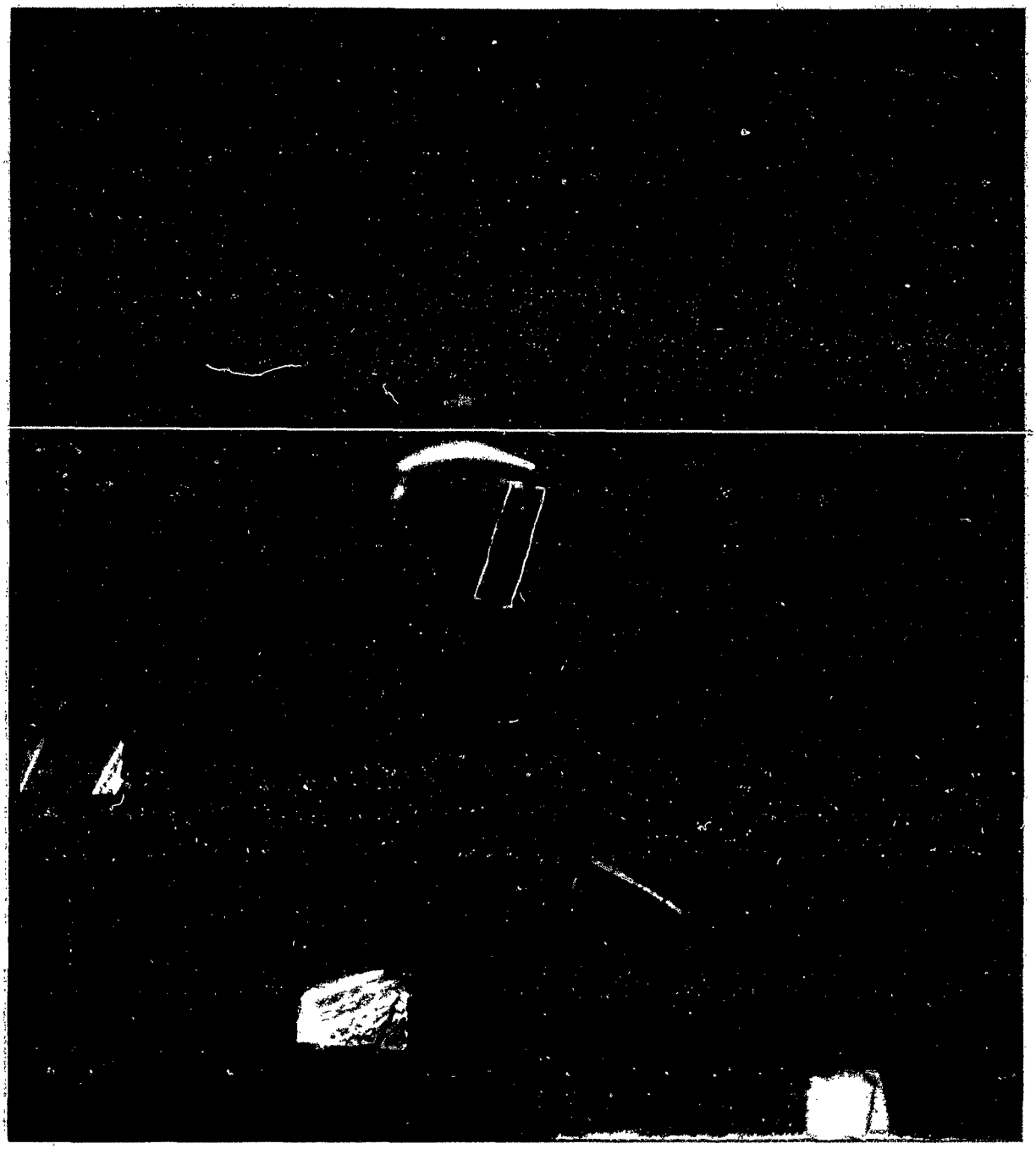

Figure 3 Schulz-Wei pattern of 1690-34 (110)[001] cross-section. [001] is perpendicular to page, (110) normal lies along vertical in page. Bracket shows misorientation of $\sim 50'$. White lines on image are characteristic radiation from tungsten target.

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FINAL TECHNICAL REPORT

ROLLED SINGLE CRYSTAL TUNGSTEN SHEET

Prepared for: Linde Company
a Division of
Union Carbide Company

Prepared by: L. Raymond

Under Subcontract 1 Prime Contract NOw 64-0055-c

APRIL 1965

AERONUTRONIC
DIVISION OF PHILCO CORPORATION
A SUBSIDIARY OF Ford Motor Company,
FORD ROAD/NEWPORT BEACH, CALIFORNIA

U-3092

FINAL TECHNICAL REPORT

ROLLED SINGLE CRYSTAL TUNGSTEN SHEET

ACKNOWLEDGEMENT

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ILLUSTRATIONS (Continued)

ILLUSTRATIONS (Continued)

SECTION 1

INTRODUCTION

The long-range objective of this program is to fabricate components from single crystals which retain their monocrystallinity after processing and exposure to high temperatures. This type of fabrication technology demands a strong interplay between research and production because some of the present-day metallurgical concepts must be modified. The entire program of single crystal component fabrication evolves around the concepts of (1) 100% recovery without the onset of recrystallization and (2) welding without the onset of recrystallization.

The materials application which has imposed such conditions as the use of tungsten as a suitable metal for service at temperatures in excess of 3500°F (1930°C). The problems encountered in the actual utilization of polycrystalline tungsten as a structural material have directed extensive research programs throughout the country toward alloying in order to accomplish the following:

(1) lower the ductile to brittle transition temperature such that the material is ductile at room temperature and

(2) raise the recrystallization temperature.

On the other hand, tungsten single crystals have inherent room temperature ductility. Also, if tungsten single crystals can be deformed with a minimum amount of work hardening introduced during processing, sufficient room temperature ductility should be retained. Finally, if recovery treatments can completely remove the work hardening introduced during forming, the annealed tungsten single crystals should have a recrystallization temperature equal to their melting temperature.

The immediate objectives of this program, the results of which are covered in this report, were to obtain the basic principles and feasibility studies on sheet rolling as the selected form of fabrication. This year's program was divided into four sections.

(1) The substructural changes that accompany tungsten single crystals deformed by rolling at temperatures in excess of 700°C and the effect of these substructural changes on the strength and ductility of the sheet material.

(2) The influence of recovery treatments on the substructure and eventually on the properties.

(3) The effects of recrystallization on ductility and workability.

(4) The metallurgical properties which include (a) fabricability with regard to rolling, (b) weldability with regard to electron beam welding and (c) the effect of these properties on transition temperature as measured in bending.

The technology generated from this year's research effort has advanced the state-of-the-art to the point that the subsequent program can now undertake the objective of actually fabricating a piece of hardware from a single crystal of tungsten. The most immediate application appears to be in liner materials or possibly in small prototype nozzles which must be evaluated with regard to thermal shock. With electron beam welding as a source of thermal shock, tungsten single crystals appear to be resistant to this thermal exposure. Also, substitutional solid solution tungsten single crystals are available for high temperature strength.

SECTION 2

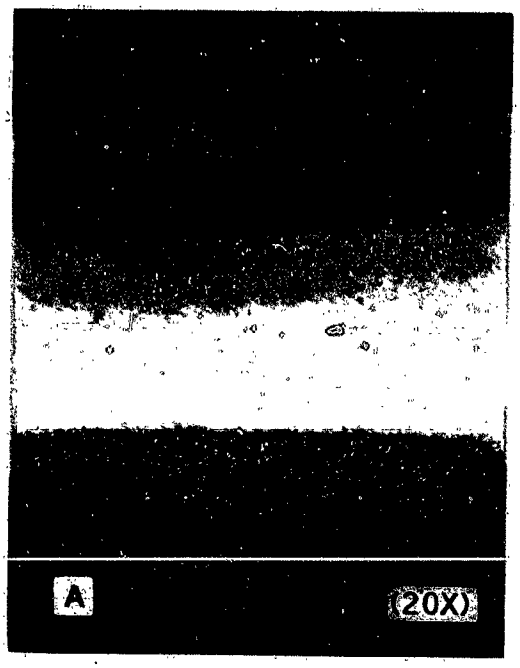
SUMMARY OF RESULTS

2.1 GENERAL

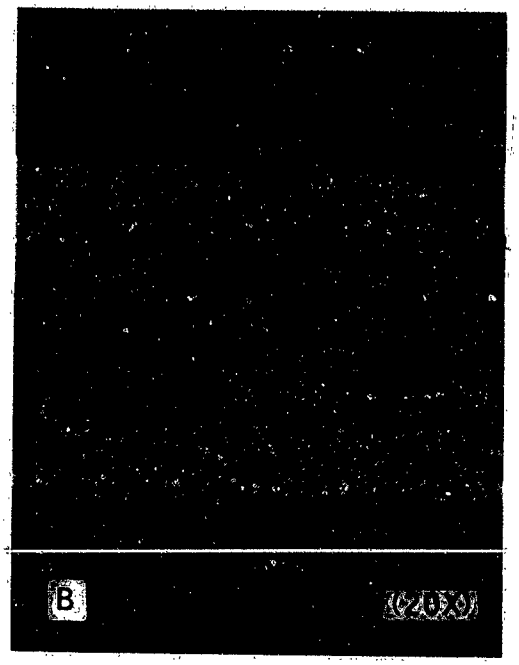
With the rapid advances in growing large single crystals from commercial purity tungsten powder using the Arc-Verneuil technique, fabrication of single-crystal sheet becomes more of a reality, owing to progress in our understanding of the associated anisotropic substructural changes that occur during rolling. Starting single-crystal billet sizes of 1½ x 3/8 x 12 in. have already been produced, with the potential final size unlimited because of the unique method of traversing the hot plasma source both reciprocally and vertically from the seed. Compared to polycrystalline material tungsten, sheet fabricated from single crystals has immediate advantages owing to the absence of grain boundaries. Single-crystal sheet can be rolled at temperatures between 700° and 1000°C; it possesses strength levels comparable to polycrystalline tungsten sheet and still has as much as 14% elongation at room temperature; it has the bend ductile-to-brittle transition temperature below room temperature; it can be welded without affecting the monocrystalline nature of the crystal; and it is free of delamination. Because of the anisotropic nature of single crystals, rolling planes and directions must be considered in selecting the orientation that best favors processing. Although still on a laboratory scale, the possibility of advancing the technology to produce large-size sheet looks promising. The more specific results of this program are itemized by sections as follows.

2.2 ON DEFORMATION BY ROLLING

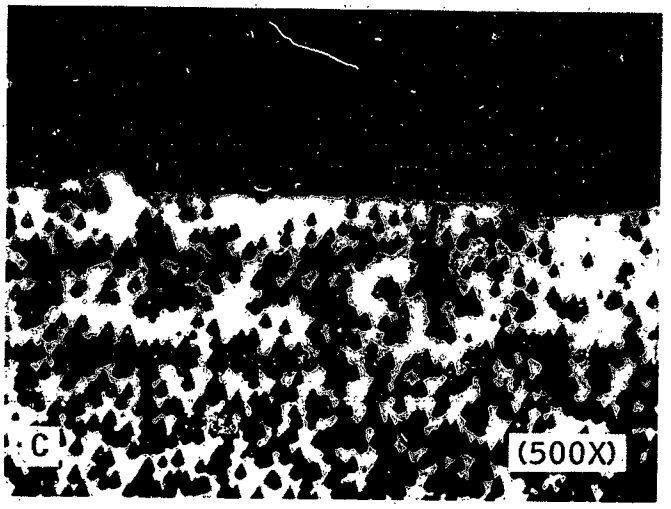
(1) Only one set of slip systems are active in tungsten crystals deformed at temperatures in excess of 700°C. This is the {112} <111>. Therefore, the substructure



AS-ROLLED



RECOVERY ON SURFACES
RECRYSTALLIZED GRAIN
IN CENTER



ETCH PITS ON RECRYSTALLIZED
CENTER SECTION

ORIENTATION APPROXIMATELY
<112> NORMAL TO PAGE

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FIGURE 19. SANDWICH STRUCTURE FORMED AFTER ONE-HALF HOUR 1600°C ANNEAL ON (110) [001] CRYSTAL REDUCED 50 PERCENT IN SIX PASSES. AS ROLLED CRYSTAL WAS DEFORMED BY SIMPLE SLIP AT SURFACE; DUPLEX IN CENTER

introduced into the crystal by rolling can be controlled by proper selection of processing variables and crystal orientation.

(2) The least work hardened condition for a given percent reduction in thickness is obtained with simple slip occurring at the rolled surfaces on conjugate planes which results in duplex slip at the center. This technique is characterized by anisotropic hardening, retention of ductility with warm working, lattice bending which is related to the contact angle, and macroscopic dimensional changes in the length but not the width of the billet.

(3) When several slip systems are operative during rolling, a greater degree of work hardening is introduced into the crystal for a given percent reduction in thickness. This rolling condition is characterized by isotropic hardening, loss of ductility with warm working, complex lattice distortions, and macroscopic dimensional changes in both the length and width of the billet.

(4) Regardless of the gross slip behavior, a highly work hardened surface condition exists due to the frictional stresses of rolling.

(5) For a reduction of 10 percent per pass, the (001) [110] and (110) [001] orientations can be deformed as discussed above in (2). In the (001) [110], the slip directions are inclined 35^{o} to the rolling direction. This angular relationship favors duplex slip of a self-aligning nature. In the (110) [001], the slip directions are inclined 55^{o} to the rolling direction. This angular relationship is not self-aligning and therefore curvature of the crystal results on rolling. The rolling procedure must be adjusted so that the conjugate system becomes operative.

(6) The duplex slip occurring in these billets does not change the original orientation of the crystals; a bending of the lattice about the transverse axis is caused by deviation of the ideal rolling conditions. The angle of bending is approximately equal to the angle of contact.

(7) In both of the above orientations, multiple slip occurred when the billet was deformed 50% in one pass.

(8) Multiple slip also occurred in the (110) [$\bar{1}$10] and (001) [100] deformed 50% by either multiple or single pass rolling techniques.

2.2.1 EFFECT ON PROPERTIES

(1) Deformation by the simple mode of slip does not effect the tensile strength although it tends to raise the yield strength.

(2) In the <110> tensile orientation, deformation by the simple mode of slip removes the yield point effect which is found in this orientation.

(3) Deformation by multiple slip increases both the yield and tensile strength at the sacrifice of ductility.

2.3 RECOVERY

(1) Complete recovery of hardness, strength and ductility was obtained in properly oriented crystals deformed 10% in one pass by annealing at temperatures in excess of 2000°C, although some lattice bending was retained.

(2) A lower annealing temperature of 1000°C produced no observable changes in hardness, etch pit density or Laue back-reflection spots.

(3) Complete softening without the onset of recrystallization was obtained for the (001) [110] crystal annealed ½ hour at 1600°C after a 50% reduction in six passes. This softening process is associated with the driving force for recrystallization i.e., recrystallization "in situ" by comparison to the recovery process occurring at slightly lower annealing temperatures.

(4) Subsequently raising the temperature after complete softening without recrystallization, i.e., 1600°C for ½ hour after 50% multiple pass reduction, produced a typical recrystallized grain structure.

2.3.1 EFFECT ON PROPERTIES

(1) Low temperature annealing (1200°C) i.e., stress relieving, has little or no effect on both the yield and tensile strengths.

(2) Low temperature annealing (1200°C) i.e., stress relieving crystals which have been reduced 50% in thickness, has an embrittling effect which is more pronounced in the crystals deformed under conditions of multiple slip than in the crystals deformed under conditions of simple or duplex slip. Since there were no detectable changes in subboundary formation, the only phenomenon which can apparently account for this embrittling behavior is the formation of interstitial atmospheres around dislocations -- even at low interstitial levels. To account for the difference in the embrittling response of crystals deformed by multiple slip versus simple slip, it must be further assumed that the interaction energy at points of dislocation intersection must be greater than along the length of the dislocation.

2.4 RECRYSTALLIZATION

(1) Recrystallization in crystals reduced 50% or less in thickness, is highly sensitive to surface conditions.

(2) With local surface conditions removed by electropolishing, the recrystallization temperature increases as the mode of slip changes from multiple slip to simple slip.

(3) Even in the crystals which are deformed by simple slip at the surface, i.e., easy glide, and duplex slip in the center, recrystallization initiates in the center of the crystal where duplex slip is apparent; suggesting therefore that duplex slip presents a more work hardened state than simple slip.

(4) For the same percent reduction by rolling, the recrystallization temperature can be varied for a given crystal orientation by modifying the rolling procedure. For example, 50% multiple pass reduction

in the (001) [110] produces a less work-hardened condition than 50% single pass reduction; therefore, the sheet deformed by multiple pass rolling will have a higher recrystallization temperature than that deformed by single pass rolling techniques.

2.4.1 EFFECT ON PROPERTIES

(1) As classically observed, recrystallization has a catastrophic embrittling effect on tungsten.

2.5 METALLURGICAL PROPERTIES

2.5.1 FABRICABILITY

(1) The initial forging, rolling, or swaging temperature of a single crystal tungsten billet is about ½ that of sintered products, i.e., 1500°F to 1800°F (800°C to 1000°C).

(2) Frictional stress in rolling can account for the difference in the rolling behavior of the (110) [001] crystal as compared to the (110) $[\bar{1}10]$. It is estimated that the frictional stress is above the same order of magnitude as the compressive stress. This accounts for the observed high dislocation density at the rolling surfaces.

(3) Macroscopic dimensional changes can be used to determine if a limiting number of slip systems are operating during rolling. If so, the billet will decrease in thickness, increase in length, but not change in width.

(4) As the specimen thickness decreases during rolling, higher roll temperatures will be necessary to maintain deformation by a simple mode of slip.

(5) The built-in dislocation grid of single crystals with known deformation behavior can be a useful tool for the investigation of the rolling process, in addition to the commonly employed quasi-isotropic polycrystalline materials.

2.5.2 WELDABILITY

Welding tungsten to itself by most conventional welding procedures results in an embrittled joint. In contrast to the typical weld joint of a recrystallized and heat affected zone found in polycrystalline material, the single crystals are completely absent of any such effects. Only by x-ray, can slight lattice misorientations be detected through the weld zone. Therefore, any embrittling effects due to welding would be absent in tungsten single crystals which have been welded by electron beam. Such behavior is tremendously advantageous and offers great flexibility into fabricating components from single crystal inasmuch as the commonly observed problems of welding are no longer applicable to single crystal sheet materials.

2.5.3 TRANSITION TEMPERATURE

The transition temperature as measured by the tensile test is below room temperature. In bending the transition temperature is found to lie between -50°F and -125°F depending on crystal orientation. The most favorable single crystal rolling orientation, (001) [110], has the lowest ductile-to-brittle transition temperature and coincidentally is the rolling texture of polycrystalline tungsten.

2.6 PUBLICATIONS

As a result of this program, two publications have resulted with a third to follow. They are:

(1) "On the Deformation of Tungsten Single Crystals by Rolling" accepted for publication March 1965, Trans. AIME.

(2) "Tungsten Single-Crystal Sheet" published in Proceedings of the 6th Structures and Materials Conference, Palm Springs, California, April 5-7, 1965.

SECTION 3

EXPERIMENTAL TECHNIQUES

3.1 SPECIMEN PREPARATION

The Linde Company supplied single crystals billets, which had a thickness of approximately 1/4 inch and a width of 3/8 inch. They were then cut with a diamond wheel into pieces of about 2 1/2 inch length and electrolytically polished in an aqueous solution of 20g NaOH per liter in order to remove a surface layer of .001 inch. The crystals were then annealed in vacuum (10^{-4} Torr) at 2400°C for 30 minutes.

After each rolling operation a test piece of about 1/4 inch long was cut off from the aft end. For the cutting operation the crystals were mounted on ceramic tiles with DeKhotinsky cement. The transverse section was then used for x-ray, hardness, and etch pit studies. On an annealed crystal, the damage introduced by cutting with a water cooled diamond wheel and subsequently grinding on 600 grit paper did not go deeper than .001 inch. A surface layer of .002 inch was therefore removed from all 1/4 inch samples by electrolytically polishing in the 2% NaOH-solution in order to eliminate all surface damage.

3.2 ROLLING OPERATION

The diameter of the rolls of a Loma Model 600 mill is 6 inches; the peripheral velocity was 500 inch/min.; the surface temperature of the rolls was about 400°-450°C. The crystals were heated before each pass for 1/2 hour in argon at 1000°C. During rolling, oxidation of the crystals in air resulted in thin layer of WO_3. This greenish-yellow oxide was removed by immersing the crystals for a few minutes in a boiling NaOH solution.

All crystals were rolled in the same direction i.e., the same end was always introduced into the rolls. The rolling program for each orientation was conducted in two ways:

(1) A reduction of 10% per pass in several passes to a total reduction in original thickness of 50%.

(2) A reduction of 50% in one pass.

The rolling was not carried beyond a final thickness of 100 mils in order to obtain round tensile bars indentical to those used by Rose, Ferris and Wulff. (1)

3.3 LAUE PHOTOGRAPHS

Initial orientation of the crystals and crystallographic changes during the rolling were observed by means of the standard Laue-Back reflection technique (30 mm distance). The pictures were generally taken in the center sections of the transverse plane with the rolling direction parallel to the incident beam. Since the effective x-ray penetration depth is less than 0.0005 inch (assuming a linear absorption coefficient of2000 cm^{-1}), complete removal of damaged surface layers was again essential.

3.4 HARDNESS MEASUREMENTS

For the hardness measurements on the transverse plane, the Vickers diamond pyramid was chosen since it is less sensitive to the orientation of the crystal than the Knoop hardness. But, the Vickers hardness still remained anisotropic. This orientation dependence was used to some extent to obtain information concerning the active slip system at room temperature. Measurements were again carried out on electrolytically polished surfaces.

3.5 ETCH PIT INVESTIGATIONS

The etch pit technique was found to be a very valuable tool in determining the mode of deformation through the thickness of the ingots, particularly at low deformations. Reduction of the voltage from 10 for polishing to 3 for etching in a 2% NaOH solution revealed subgrain boundaries and individual dislocations. The etch pits on the {100} faces have a square shape, with the sides of the squares parallel to the <100> directions. At room temperature, the etching time was about 5 to 10 seconds.

3.6 TENSILE SPECIMENS

Because of the extreme difficulty experienced in machining sub-size sheet tensile specimens and the further difficulty in gripping the specimens during testing, the room temperature mechanical properties were measured on round tensile bars ground from the rolled sheet. The size was chosen to be exactly the same as that used by Rose, Ferris and Wulff[1] in studying annealed single crystal properties, the results of which will be used for comparison.

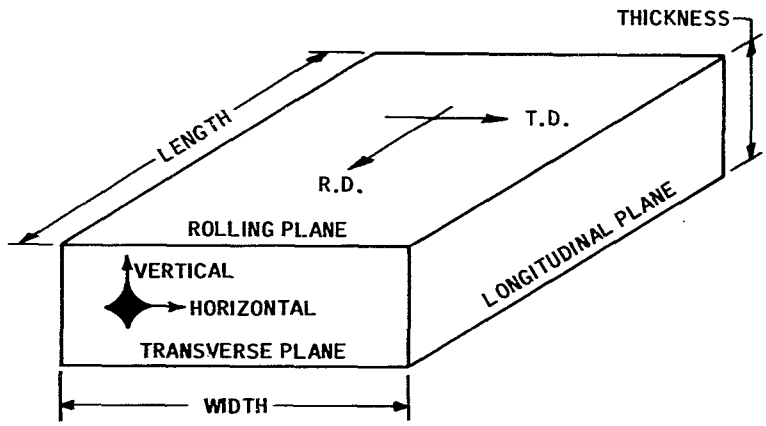
Testing was done on an Instron at a strain rate of 0.025 per minute. The gage length was about 0.8 inch with a $0.050 \pm .002$ inch diameter. At least .004 inch was previously removed from the diameter by polishing in order to remove all of the surface damage introduced by grinding the specimens to their final shape. All specimens were successfully ground and tested with no premature breakage during either operation.

SECTION 4

RESULTS

4.1 ON DEFORMATION BY ROLLING

Throughout this investigation, the different faces of the crystal and their related directions were defined as illustrated in Figure 1. In terms of their rolling plane and rolling direction, the billets had the following orientations: (110) [001], (110) [$\bar{1}$10], (001) [1$\bar{1}$0] and (001) [$\bar{1}$00]. The actual orientation of all crystals used in this investigation was within 5° of the ideal orientations.



R.D. = ROLLING DIRECTION
T.D. = TRANSVERSE DIRECTION

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FIGURE 1. FACES AND RELATED DIRECTIONS OF ROLLED SINGLE CRYSTAL BILLET

4.1.1 MACROSCOPIC DIMENSIONAL CHANGES

When a single crystal is deformed by rolling, the macroscopic dimensional changes of the billet depend on the relative orientation of the active slip systems. When the crystal orientation is such that only one set of primary slip systems are operative, elongation occurs only in one direction. Once secondary slip systems become operative, dimensional changes occur in all directions with such effects as "barrelling" often accompanying this complex mode of deformation. Therefore, macroscopic dimensional changes reflect well-defined internal changes which, in turn, establish the final mechanical properties of the rolled sheet.

From the data in Table I, plotted in Figure 2, it is observed that two favorable orientations can tolerate from 60 to 70 percent reduction in thickness with little or no widening of the billet. This would be accompanied by about 200% elongation i.e., the length of the billet would have been doubled. The less favorable orientations experience about as much widening as they do percent reduction in thickness; therefore, for about 80% in comparison to 200% by the simple mode of deformation. All four crystals were deformed on a nominal 10 percent reduction per pass in multiple-passes. Under these conditions, the two favorably orientated crystals $\{(001)\ [110],\ (110)\ [001]\}$ deformed by simple slip of two conjugate slip planes; whereas the two unfavorable oriented crystals $\{(110)\ [\bar{1}10],\ (100)\ [001]\}$ deformed by multiple slip.

If the processing variables are not selected properly, the favorably oriented crystals could also have been deformed by multiple slip. An example is obtained by deforming the two favorably oriented crystals 53% in a single pass. The results are superimposed on the plot in Figure 2. It is observed that the macroscopic dimensional changes of the billet are solely a function of the rolling plane, independent of the rolling direction under these severe conditions of deformation. The (110) [001] widens in a single 53% pass as much as the (110) $[\bar{1}10]$ widens in 53% by 10% multiple-pass rolling. Likewise, the (001) [110] widens in a single 53% pass as much as the (100) [001] widens in 53% reduction of thickness by 10% multiple-pass rolling -- suggesting that identical slip systems are operative in each comparative case, independent of rolling direction.

Only the thickness and width were measured after each pass since 0.25 inch sample sections were removed from the aft ends for microscopic examination. Therefore, the length was calculated from the relationship:

$$(1 + \epsilon_l)\ (1 + \epsilon_w)\ (1 + \epsilon_t) = 0$$

where $\epsilon_{l,w,t}$ = nominal strain in the length, width, or thickness.

TABLE 1

DIMENSIONAL CHANGES OF BILLETS DURING ROLLING

(110) [001]		(001) [110]		(100) [001]		(110) [$\bar{1}$10]	
ϵ_t	ϵ_w	ϵ_t	ϵ_w	ϵ_t	ϵ_w	ϵ_t	ϵ_w
12	0	11	0	10	5	7	7
22	0	22	0	17	9	20	19
27	0	30	1	27	14	28	24
35	0	37	1	31	16	37	31
44	0	44	2	36	20	91	86
53	0	55	2	40	23		
60	1	64	5	92	72		
69	3	71	7				
77	14	80	14				
83	30	85	28				
86	39	88	36				

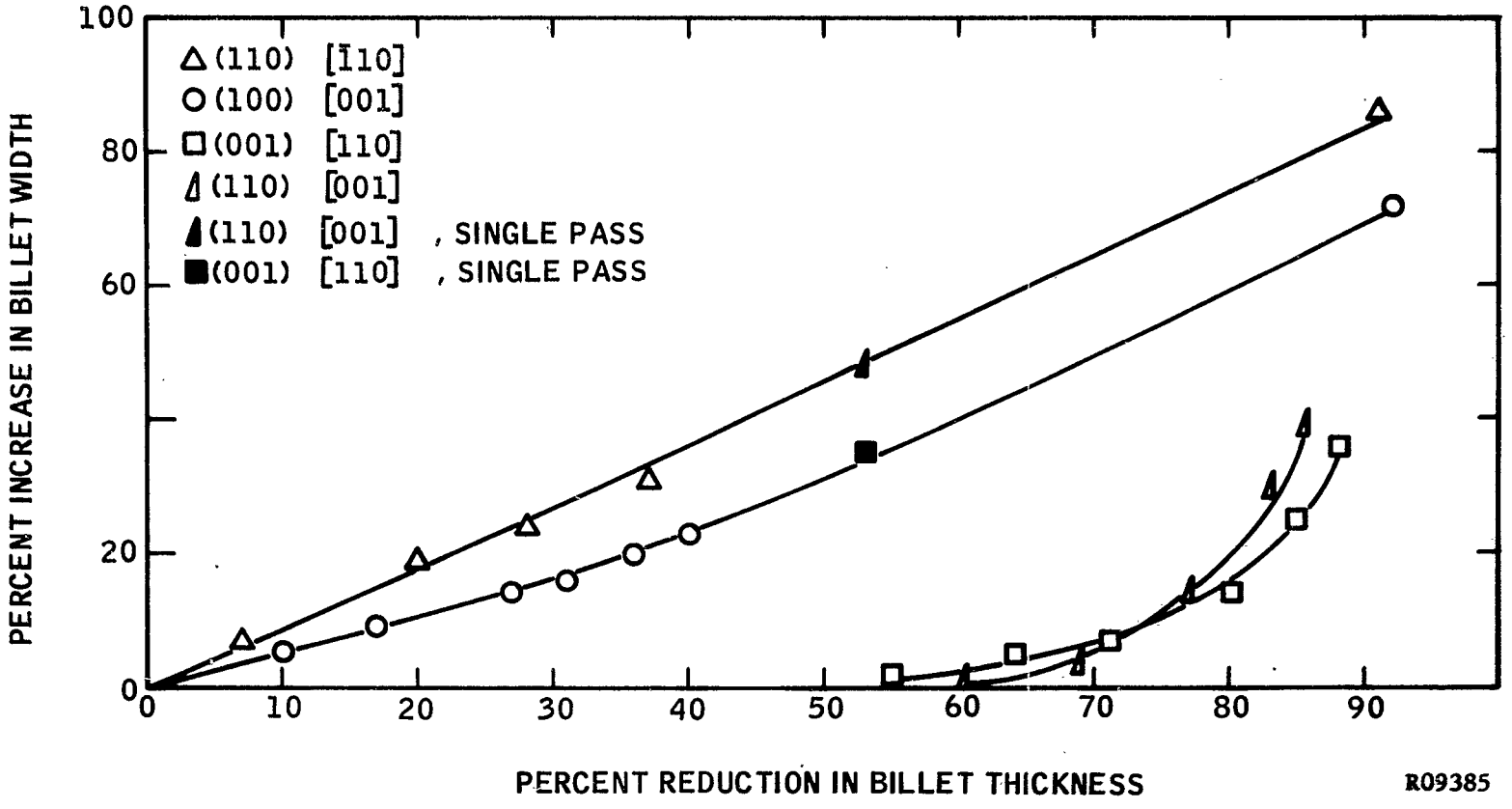


FIGURE 2. MACROSCOPIC DIMENSIONAL CHANGES ACCOMPANYING ROLLED SINGLE CRYSTAL BILLET

4.1.2 HARDNESS ANISOTROPY

Because of its monocrystallinity, the hardness measurements taken on a specific plane of a tungsten single crystal are directional or anisotropic. With the Knoop hardness tester, this anisotropy was found to account for about 15% variation in readings which is in agreement with another investigation[2] conducted on Columbium single crystals. With the Vickers hardness tester, anisotropy account for about 6% variation in readings. The Knoop indenter was also found to be much more sensitive than the Vickers to local surface disturbances such as slight curvatures, surface roughness, etc., which often resulted in extremely erratic hardness measurements. For these reasons, the Vickers hardness measurements were selected as a means of recording the hardness changes accompanying deformation by rolling.

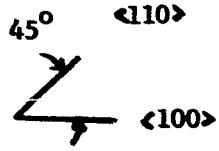
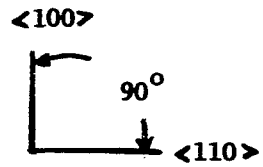
The Vickers hardness indentation is made with an equiaxed, diamond-shaped point. Hence the difference in the length of each of the perpendicular axes gives a hardness measurement in two directions and hence a measurement of the anisotropic nature of the crystal. In comparing (Fig 3A) Vickers and Knoop hardnesses, it must be remembered that the length of the long axis of the Knoop indentor represents the hardness in the direction of the short axis. Table II lists the hardness of annealed tungsten single crystals with indentations made on either the {100} or {110} type planes and the <100> or <110> directions. On the {100}, the <110> and <100> are 45° to each other; whereas on the {110}, the <110> and <100> are 90° to each other.

Examining Table II, the hardness measurement is detected to be only slightly sensitive to the load (about 2% deviation). The shape of the indentation is illustrated in Figure 3 to be sensitive to the direction even on the same plane. Two indentations, taken on the (100) plane of an annealed crystal only rotated 45° against each other, have a diamond shape with concave sides versus a diamond shape with convex sides. The concavities in the diamond shape are normally observed in annealed materials. The barrelling effect of the diamond shape is generally associated with a work hardened material. In the annealed tungsten single crystal, both shapes appear.

On the (100) plane, the indentation axes are equal for the annealed crystal in both the <100> and <110> directions (Figure 3). For the (110) plane, a single indentation gives the hardness in both the <100> and <110>directions because they are perpendicular to each other. In this latter case, the axes are unequal in length with the <110> direction displaying a higher hardness number (about 363 VHN versus 341 VHN).

TABLE II

HARDNESS ANISOTROPY OF ANNEALED TUNGSTEN CRYSTALS

{100} Plane	<100> Direction	Shape of Indentation
	339 VHN (1 Kg)	
	342 VHN (20 Kg)	Equiaxed and barrel-shaped
	349 VHN (50 Kg)	
	<110> Direction	
	325 VHN (1 Kg)	
	325 VHN (20 Kg)	Equiaxed and concave-shaped
	330 VHN (50 Kg)	
{110} Plane	**<100> Direction**	
	343 VHN (1 Kg)	
	335 VHN (20 Kg)	Unequal axes and slightly concave
	347 VHN (50 Kg)	
	<110> Direction	
	358 VHN (1 Kg)	
	362 VHN (20 Kg)	Unequal axes and slightly concave
	368 VHN (50 Kg)	

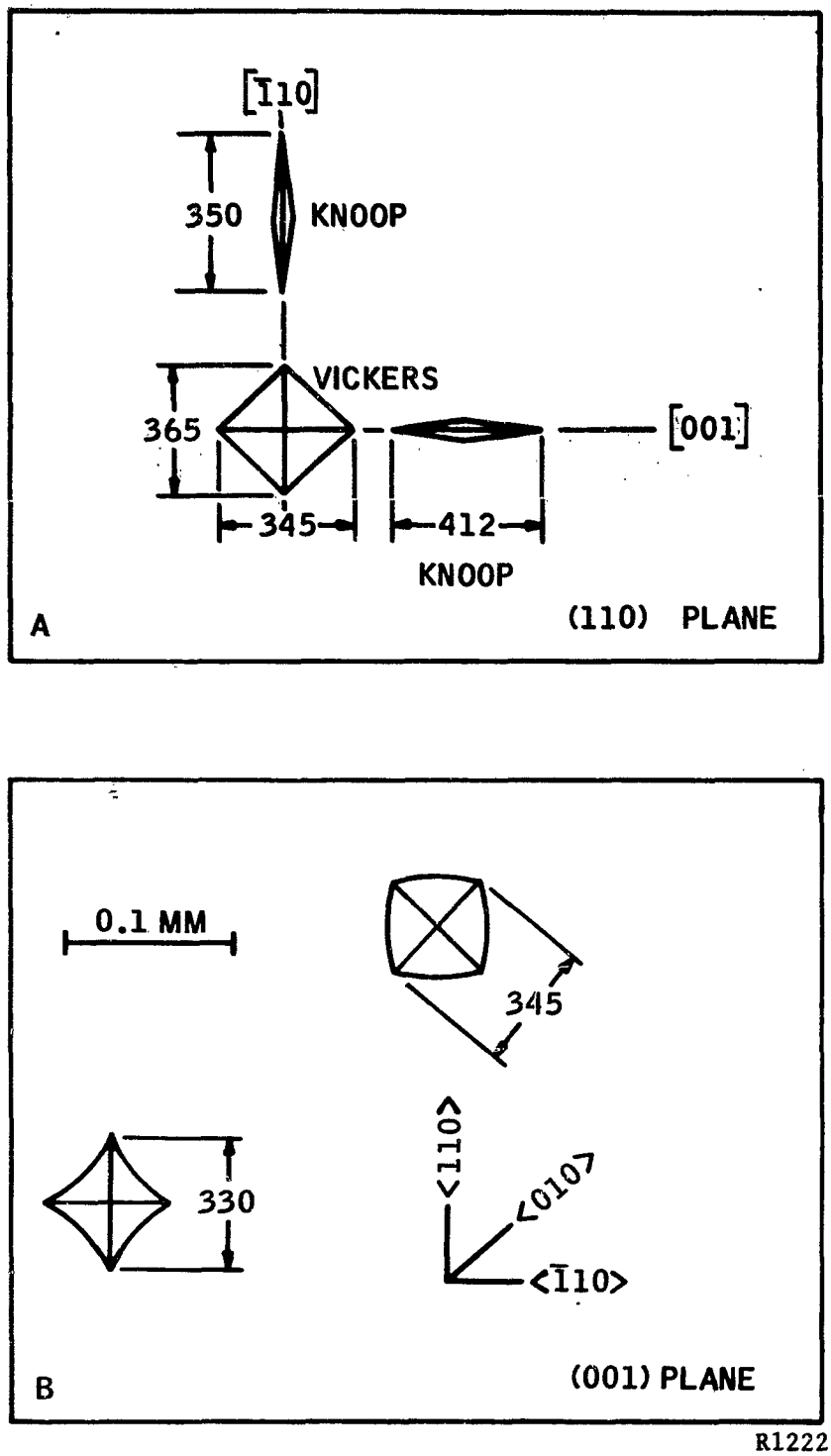


FIGURE 3. ANISOTROPY OF VICKERS HARDNESS ON THE (110) AND (001) PLANES IN AN ANNEALED CRYSTAL

4.1.3 HARDENING ANISOTROPY

An annealed tungsten single crystal is inherently anisotropic. Deformation of the crystal by rolling can also harden the lattice in a directional or anisotropic manner, especially if only one or two slip systems are operative. Therefore, anisotropic hardening is differentiated from hardness measurements which reflect the inherent anisotropy of the crystal lattice.

The hardness measured on both the transverse plane and rolling plane as a function of percent reduction in thickness shows that some crystals have more directionality in hardening than others. Hardness measurements were taken on all four crystal orientations selected for this investigation. The combinations of hardness plane and hardness direction comply to the combination of planes and directions given in Table II. Variations in hardness readings across the thickness of the specimens were not significant enough to draw any conclusions about a hardness profile through the thickness. The average of all values obtained across the thickness are plotted in Figure 4 against the percent reduction of original thickness.

In the (100) [001] and (001) [110] crystals, which have elongated without billet widening, the hardening is seen to be highly anisotropic--being greater in the horizontal than in the vertical direction. For the (001) [100] crystal, the hardening is essentially isotropic throughout the deformation process. For the remaining (110) [$\bar{1}$10] crystal, hardening appears to be slightly anisotropic at the early stages of deformation; but after 50% reduction, the hardness values converge.

In order to more clearly demonstrate anisotropic hardening over large amounts of deformation in the two crystal orientations which did not display billet widening, the measured anisotropic hardness values of the annealed crystals are subtracted from the measured values of the deformed crystals. Figure 5 is the result. From this plot, it is observed that hardening is greater in the transverse direction of the rolling plane than in the rolling directions; also, hardening is greater in the horizontal direction of the transverse plane than in the vertical direction.

4.1.4 LAUE BACK-REFLECTION STUDIES

The most significant results of the Laue back reflection photographs was that two crystals, the (110) [001] and the (001) [$\bar{1}$10] orientations, maintained their original orientation up to 50% multiple-pass reduction. Only streaks, extending symmetrically about the Laue spots, were observed. At high deformations the streaks appeared more washed out and slightly increased in length. See Figure 6 and Table III. The results indicate

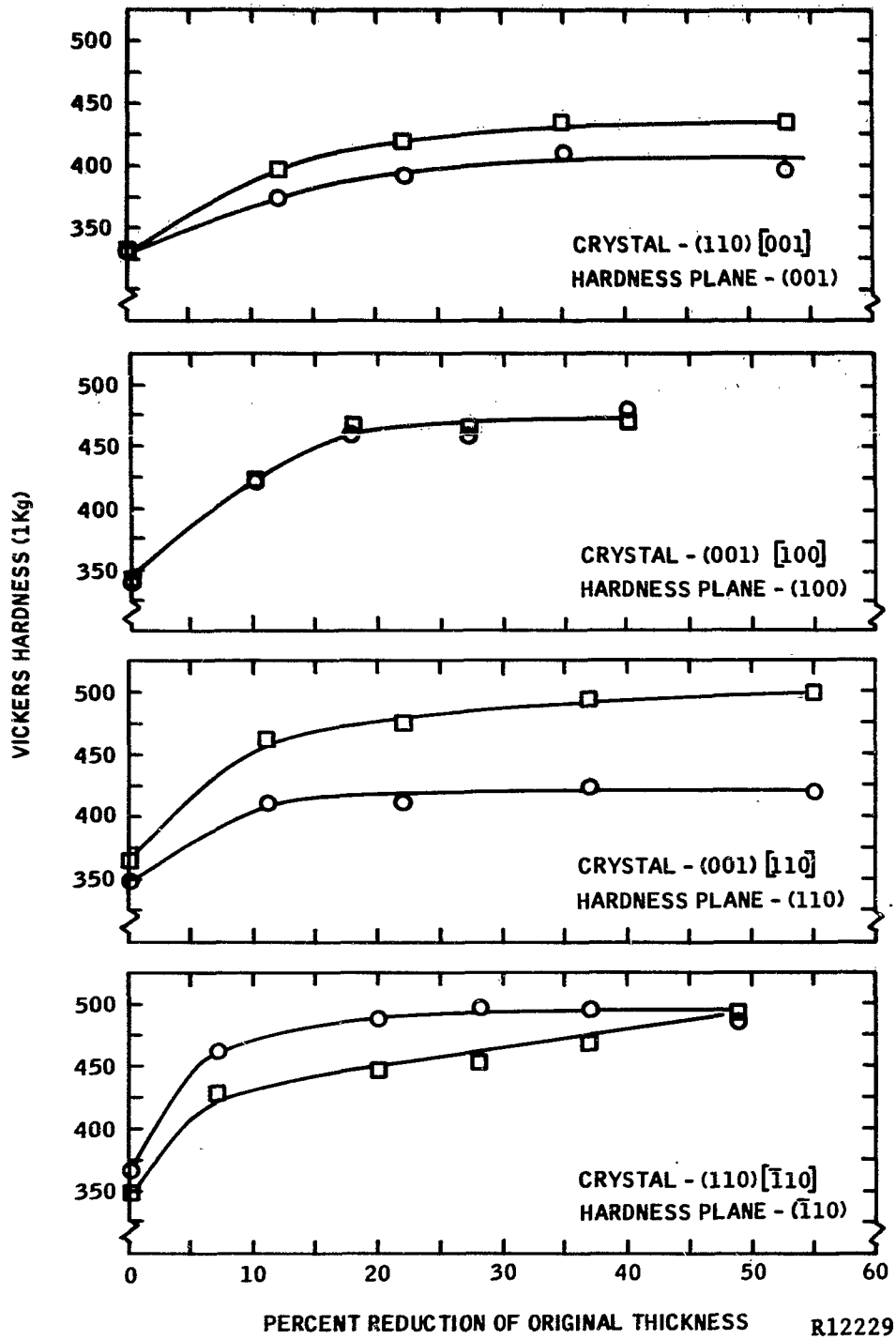


FIGURE 4. VICKERS HARDNESS AS A FUNCTION OF ORIENTATION AND DEFORMATION: [□ - HORIZONTAL, ○ - VERTICAL]

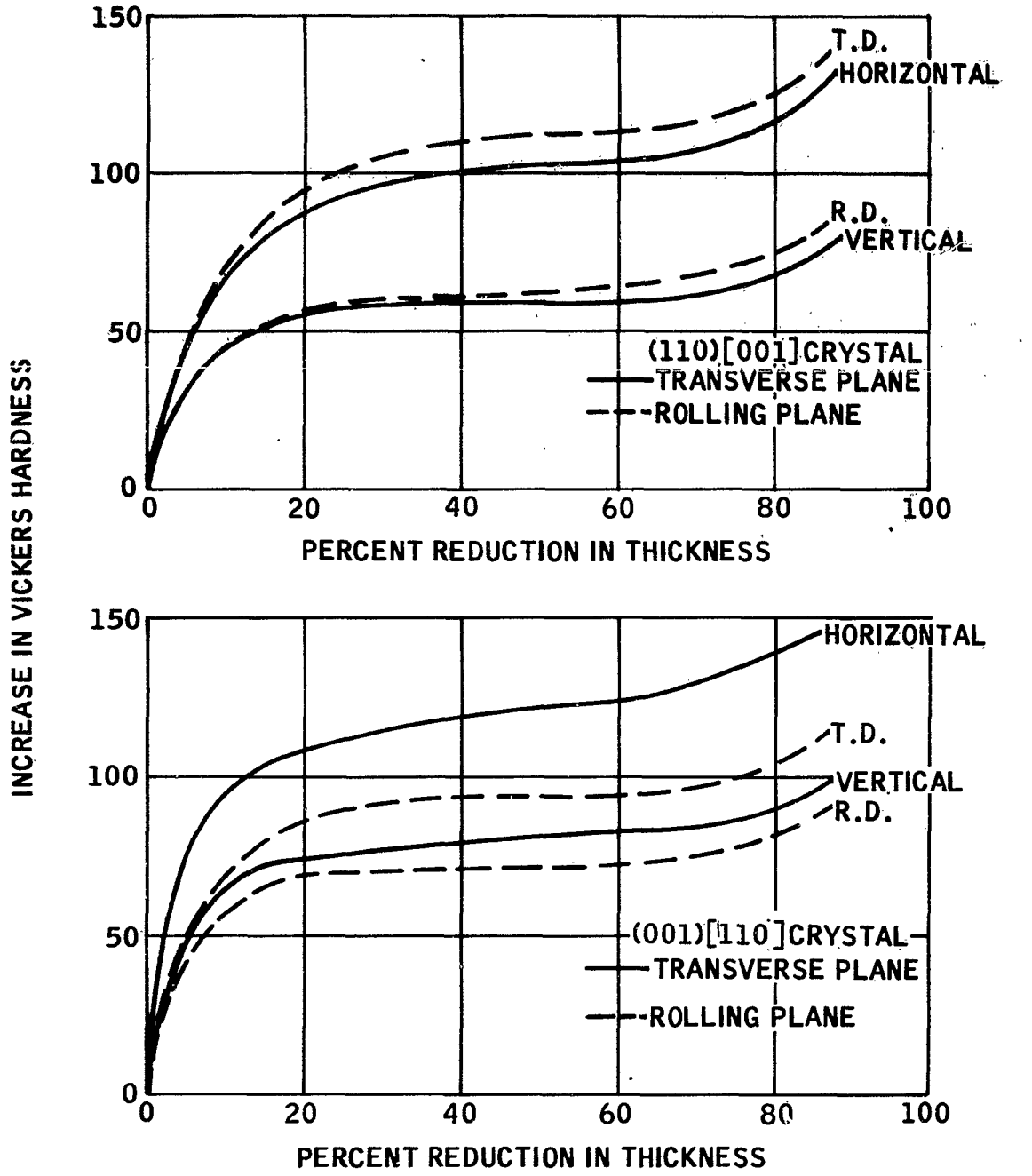


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FIGURE 5. HARDENING ANISOTROPY IN TUNGSTEN CRYSTALS. HARDNESS VALUES OF THE ANNEALED CRYSTALS ARE SUBTRACTED FROM HARDNESS VALUES OF THE ROLLED CRYSTALS.

TABLE III: STREAK LENGTH AS A FUNCTION OF THE NUMBER OF PASSES

Crystal Orientation: (001) [110]

Thickness Reduction	Streak Length
0 to 11%	8^{o}
11 to 22%	12^{o}
22 to 30%	15^{o}

TABLE IV: BENDING ANGLE AS A FUNCTION OF CONTACT ANGLE

Crystal Orientation: (110) [001]

Thickness Reduction in One Pass	Contact Angle	Bending Angle
7 %	4.2^{o}	2^{o}
12.5%	5.7^{o}	5^{o}
20 %	7.3^{o}	8^{o}
53 %	12^{o}	Debye rings

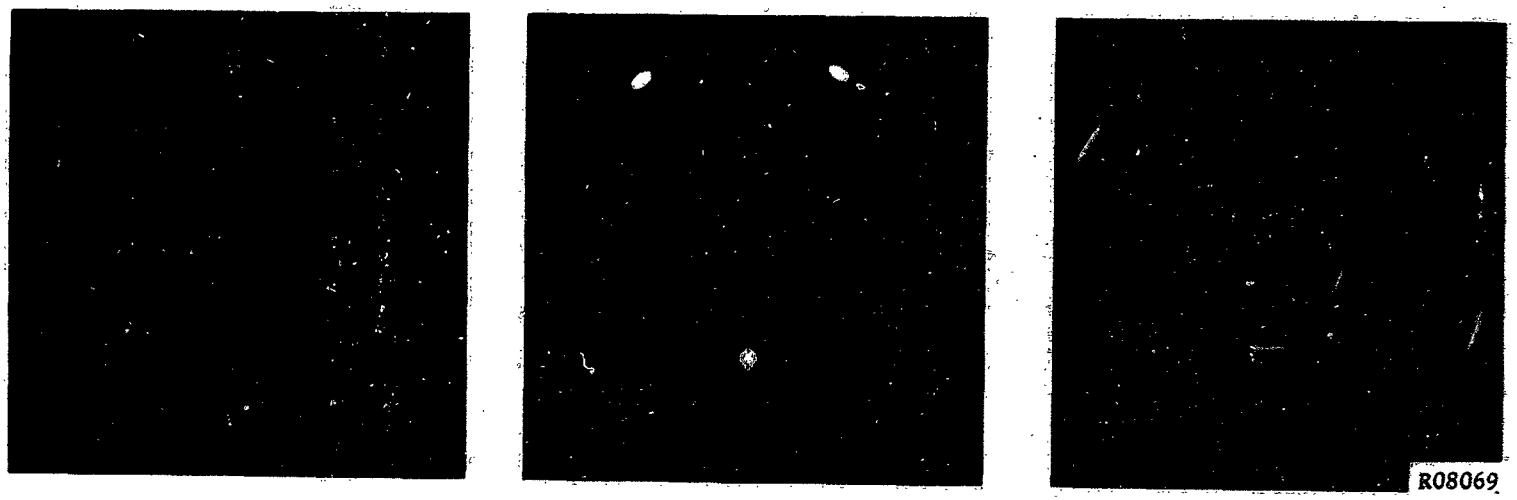


FIGURE 6. INFLUENCE OF REDUCTION PER PASS ON THE CRYSTALLOGRAPHIC CHANGES FOR (110) [001] CRYSTAL

that only a simple bending of the crystals about the transverse axis had occurred. Heating the test specimens to 1000°C for 30 minutes prior to the subsequent pass did not alter the substructure as verified by no detectable changes in the Laue photographs or hardness measurements after heat treatment.

While for both crystal orientations the streaks in the center section of the transverse plane were symmetrical about the Laue spots, photographs taken close to the top and bottom surfaces showed streaks of half this length, extending from the original Laue spots only toward the center of the billet. The contact angle of rolling, which is given by $\cos\alpha = (1 - h/2r)$ where r = radius of the rolls and h = total reduction in thickness, was found to be related to the angle of lattice bending close to the surfaces. Within the limits of error, the relationship is shown in Table IV.

For the same two crystal orientations, 50% deformation in one pass, which corresponds to a contact angle of 12°, produces Debye-rings from the characteristic radiation (Fig. 6). These Laue patterns indicate bending of the lattice about several axes due to a more severe loading condition. For the two remaining (110) $[\bar{1}10]$ and the (001) [100] orientations, Debye rings were observed in all cases after 50% reduction--single as well as multiple-pass reduction (Fig. 7). After the first pass on the (110) $[\bar{1}10]$ crystal, Laue spots on the transverse plane were only slightly distorted; but on the longitudinal plane, unidirectional streaks appeared, indicating only lattice bending about the rolling direction. This observation is in agreement with the dimensional changes recorded on the billet after the first pass. The second pass immediately resulted in distortions in several directions with Debye rings forming during the early stages of deformation. This is in agreement with the macroscopic dimensional changes which exhibit billet widening with no elongation after the first pass (Table I). After subsequent passes, both elongation and widening of the billet occurred (Table I).

4.1.5 ETCH PIT STUDIES

Etch pit studies were carried out for the (110) [001] and (001) [110] crystals deformed by multiple-pass rolling. Faces of the {100} type were always used. For annealed crystals, the dislocation density was about $10^5/cm^2$ within the subgrains. After the first rolling pass, the density went up sharply with the subgrain boundaries no longer apparent. With continued deformation, etch pits on the transverse and rolling planes formed lines parallel to the transverse direction; while on {100} faces at 45° to the transverse and rolling planes, a criss-cross pattern was revealed. (Fig. 8). This alignment was observed from 10% to 50% deformation. The criss-cross pattern appeared only in the center section of the crystals; toward the top surface only one direction appeared while close to the bottom surface the other direction predominated as indicated

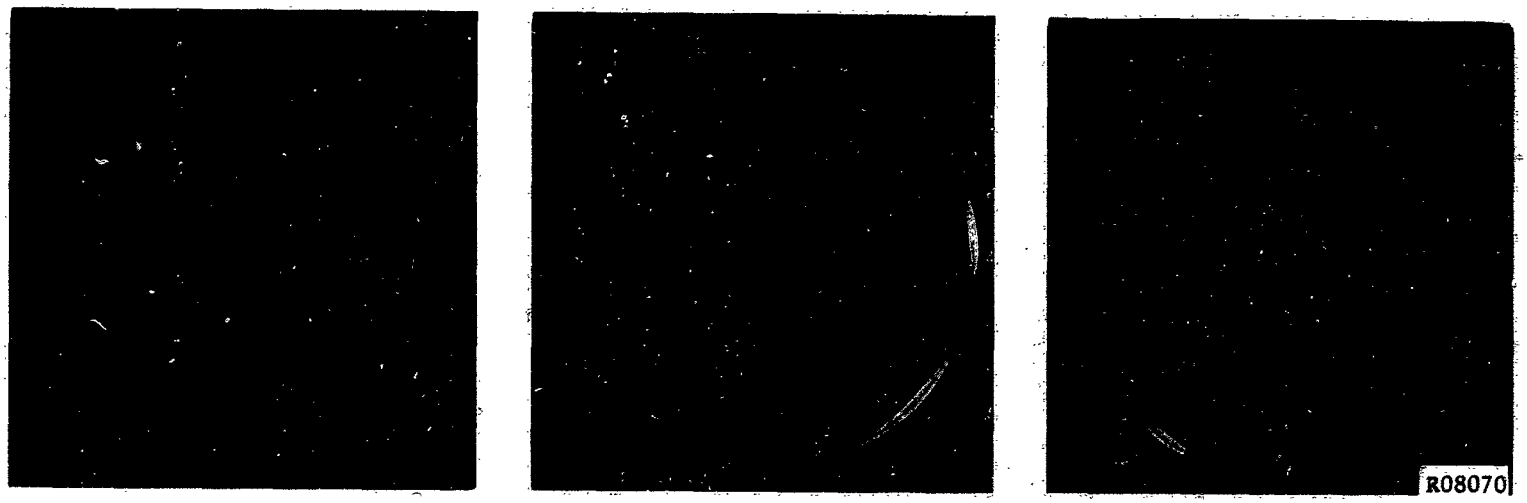


FIGURE 7. INFLUENCE OF REDUCTION PER PASS ON THE CRYSTALLOGRAPHIC CHANGES FOR (110) $[\bar{1}10]$ CRYSTAL

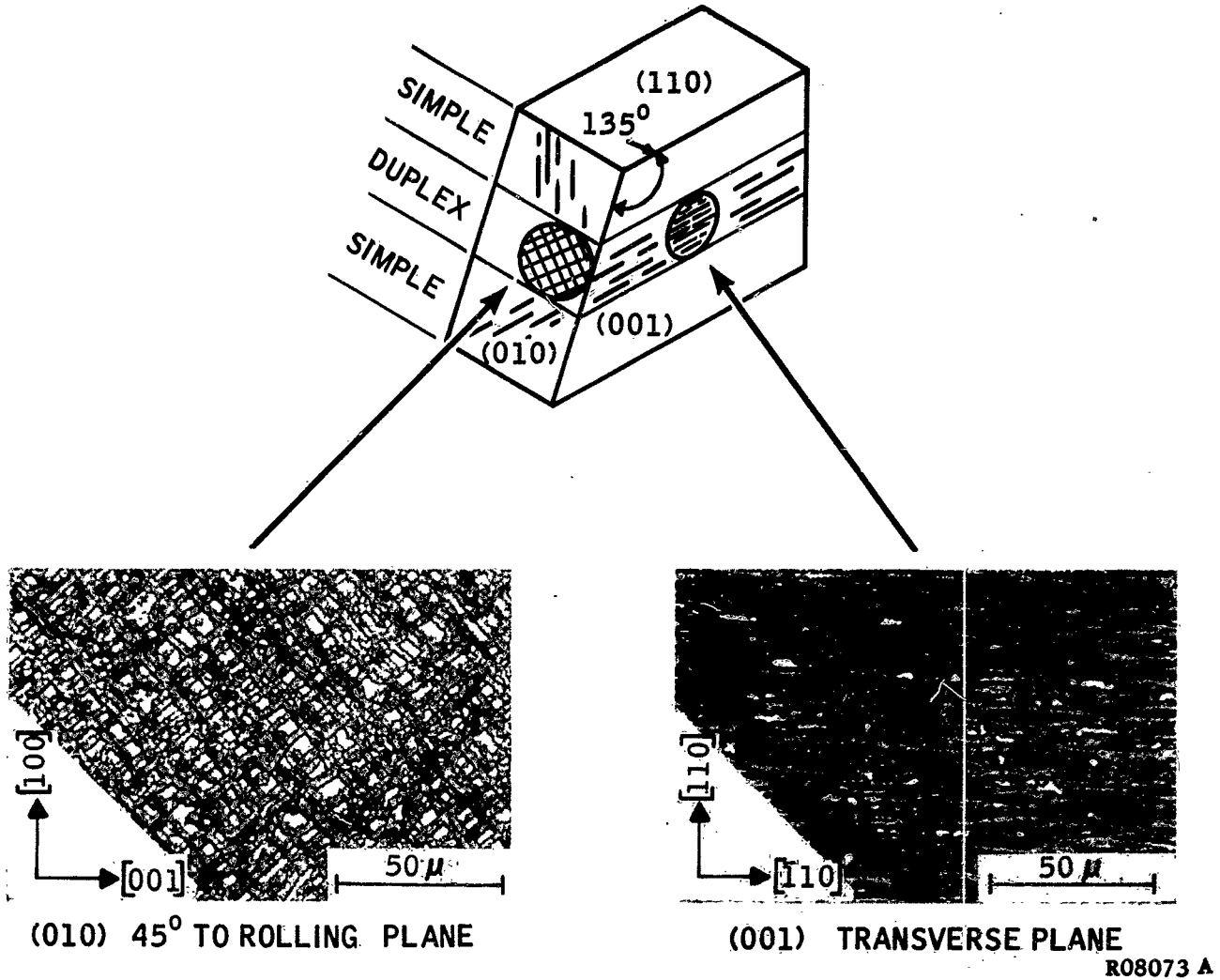


FIGURE 8. ETCH PIT PATTERNS IN THE CENTER OF THE (001) TRANSVERSE PLANE AND THE (010) PLANE AT 45° TO THE ROLLING PLANE FOR THE (110) [001] CRYSTAL DEFORMED 20% IN TWO PASSES

in Figure 9. These etch pit observations are in agreement with the Laue back-reflection observations.

A trace analysis of the planes formed by the etch pit lines showed that they were within 5° of two conjugate {112} planes, which have their normals coplanar with both the rolling direction and the normal to the rolling plane. For the (110) [001] crystal, the {112} normals form an angle $\phi = 35^{\circ}$ with the rolling direction. For the (001) [110] crystal, $\phi = 55^{\circ}$. In comparing the two orientations close to either the top or bottom surface where slip had occurred on only one system, the active slip plane of the conjugate pair was exactly the opposite, as shown schematically in Fig. 9.

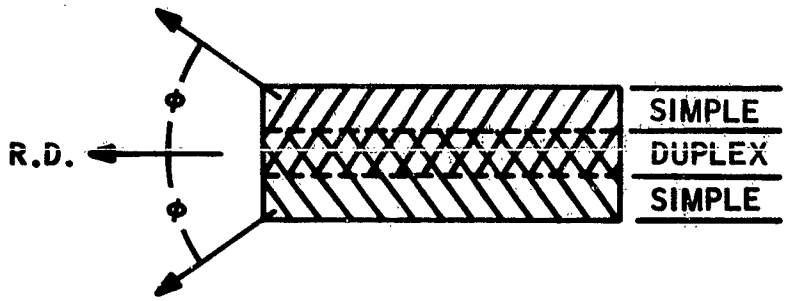
4.1.6 EFFECT ON TENSILE PROPERTIES

The tensile axis of specimens machined from the rolled sheet was coincident with the rolling direction. Two tensile directions are common to all four crystal orientations used in this investigation. Figures 10 and 11 show the results with annealed stress-strain data taken from another report.(1)

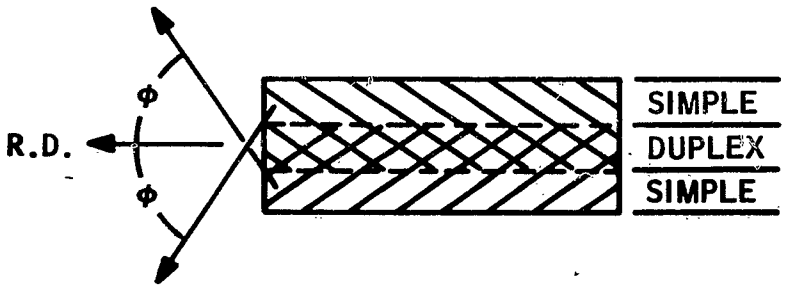
To be observed is the elimination of the yield point effect in the <110> tensile orientation when deformation occurs by simple and duplex slip. The lower yield strength is raised to the value of the upper yield point. In fact, this is the most pronounced effect for deformation by duplex slip for neither the tensile strength nor the ductility are affected in either tensile orientation. The specimens with maximum ductility (14%) were the (110) [001] and (001) [110] crystals which had all been deformed by duplex slip (Figs. 10 and 11). Fifteen percent elongation at room temperature has been reported(3) for another annealed single crystals.

The mode of fracture of the rolled crystals with high ductility was then same as reported for the annealed crystals.(4) See Figure 12. The [001] crystal exhibited uniform reduction of area, with cleavage on the (001) plane. The [110] crystals showed wedge-type fracture with an elliptical cross-section. No measurable reduction of thickness occurred along the $[\bar{1}10]$ direction, which was the major axis of the ellipse.

For deformation by multiple slip, an increase occurs in yield and tensile strength with a pronounced decrease in ductility. The (110) $[\bar{1}10]$ crystal orientation deformed 57% in one pass (Figure 10) had a maximum tensile strength of 213,000 psi; although it also had the minimum measured ductility, 2.5%. It becomes immediately apparent that deformation by multiple slip is detrimental to the ductility whether it is by multiple or single pass rolling techniques.



(110) [001] CRYSTAL: $\Phi = 35^{\circ}$, $\lambda = 55^{\circ}$



(001) [110] CRYSTAL: $\Phi = 55^{\circ}$, $\lambda = 35^{\circ}$

Φ = ANGLE BETWEEN R.D. AND NORMAL TO SLIP PLANE

λ = ANGLE BETWEEN R.D. AND SLIP DIRECTION

R12230

FIGURE 9. ORIENTATION OF SLIP PLANE TRACES NEAR THE SURFACES FOR THE (110) [001] AND (001) [110] CRYSTALS

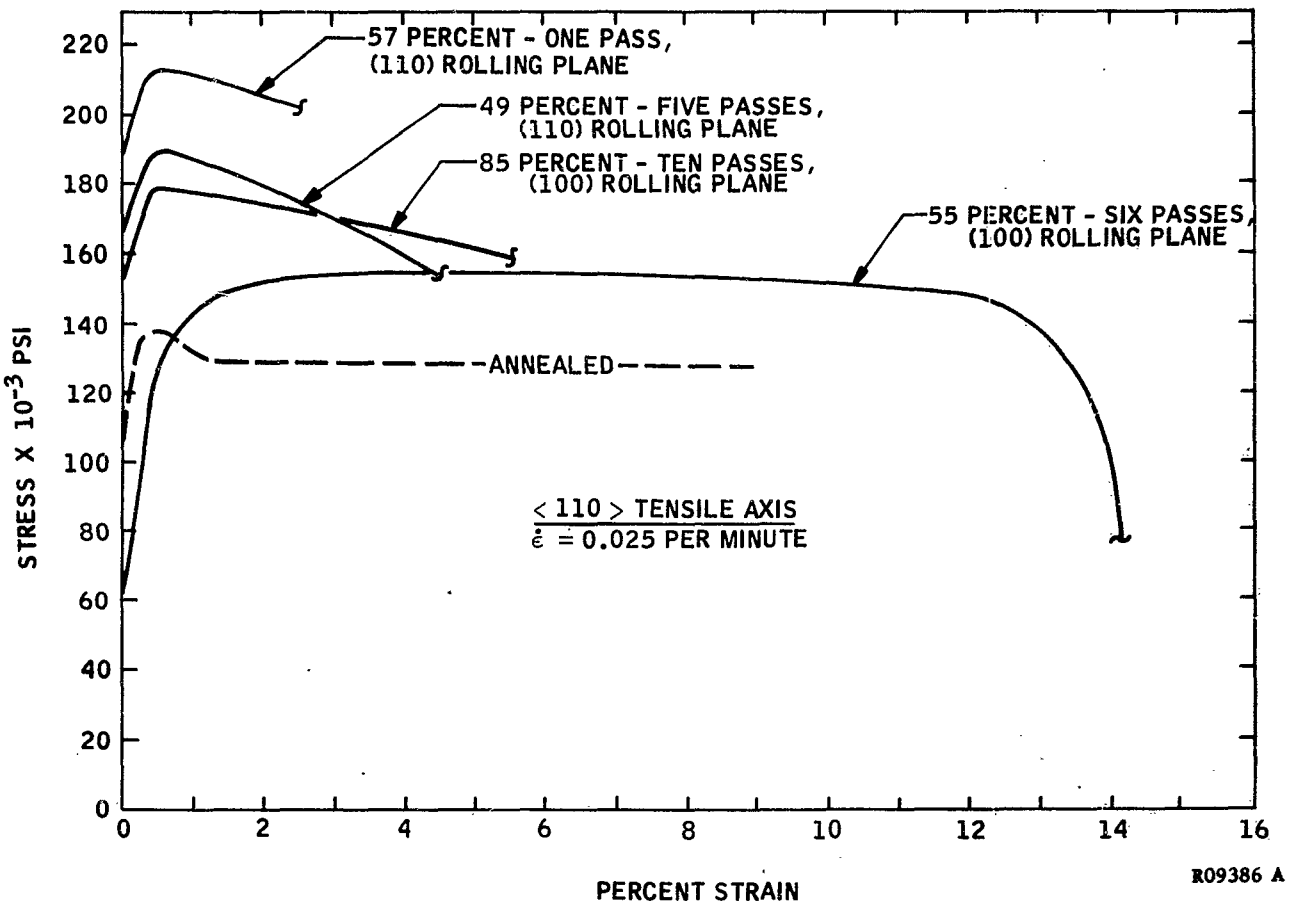


FIGURE 10. INFLUENCE OF ROLLING PLANE AND PERCENT REDUCTION ON THE ROOM TEMPERATURE TENSILE PROPERTIES IN THE < 110 > DIRECTION. TENSILE AXIS TAKEN IN ROLLING DIRECTION.

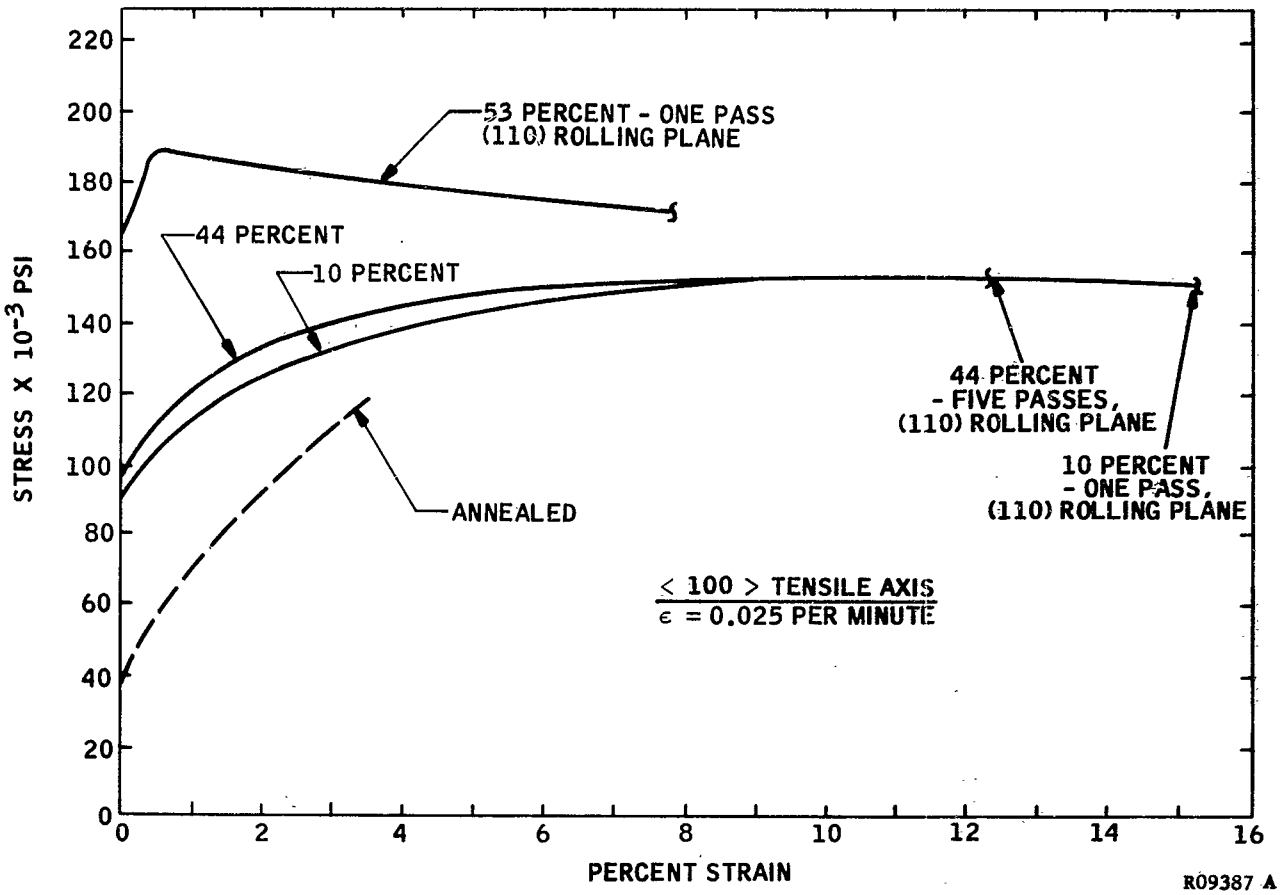


FIGURE 11. INFLUENCE OF ROLLING PLANE AND PERCENT REDUCTION ON THE ROOM TEMPERATURE TENSILE PROPERTIES IN THE < 100 >DIRECTION. TENSILE AXIS TAKEN IN ROLLING DIRECTION.

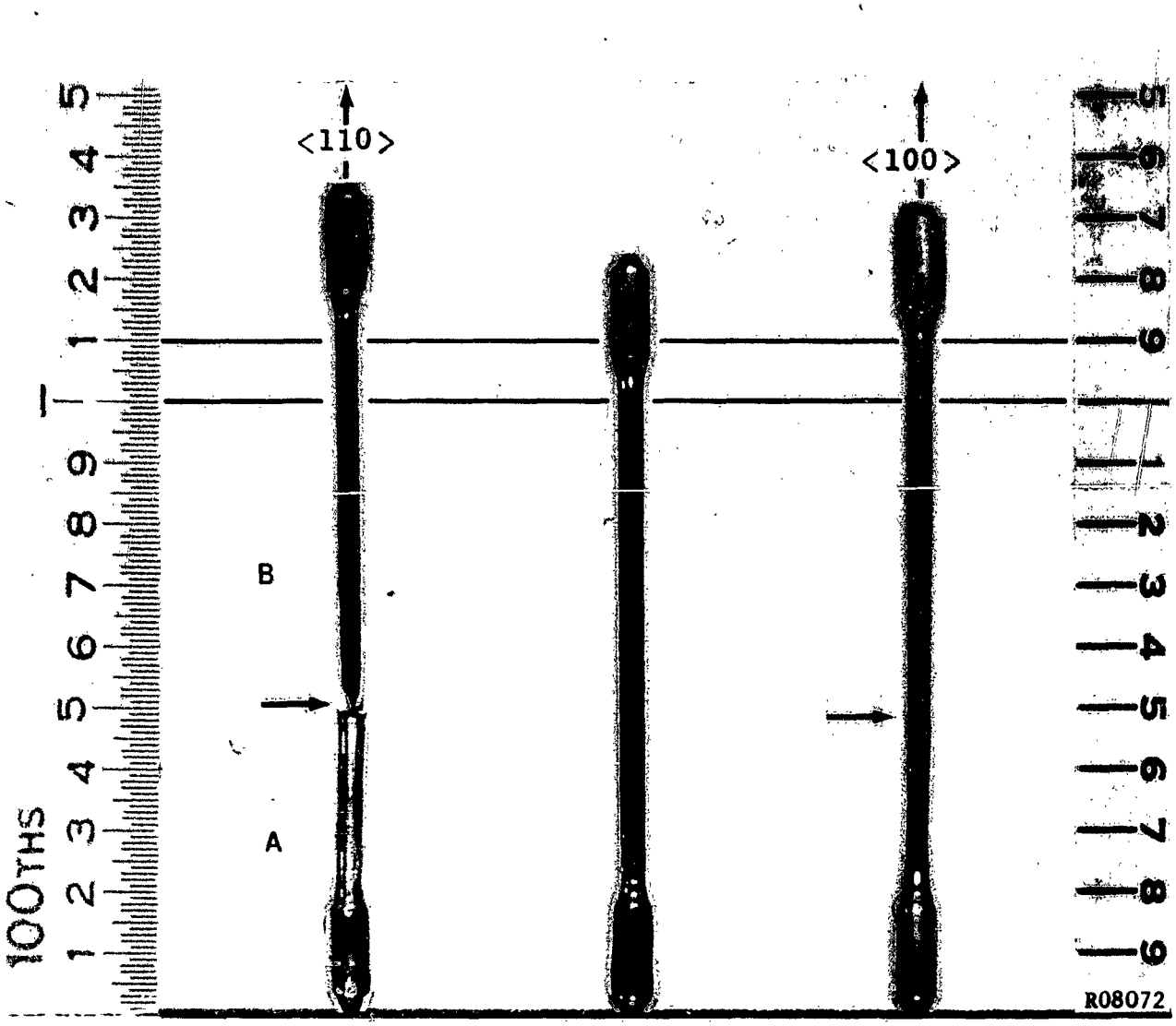


FIGURE 12. TENSILE BARS FROM W SINGLE CRYSTALS ROLLED 50 PERCENT AT 1000°C IN 5 PASSES ILLUSTRATING ORIENTATION DEPENDENCE OF FRACTURE AT ROOM TEMPERATURE. A ROTATED 90° FROM B

4.2 RECOVERY

At the completion of last year's program it was possible to utilize multiple roll-anneal techniques at 3% reduction per pass followed by a subsequent recovery treatment to retain a completely recovered or annealed substructure. The goal of this year's effort was to increase the feasibility of the program by increasing the percent reduction per pass to a possible 10% and still use the same sequence of treatments of rolling and annealing in order to retain a completely annealed or recovered crystal. This objective can only be accomplished by a more thorough understanding of substructural changes attending deformation and their subsequent response to annealing treatments.

The results presented in the previous section have clearly demonstrated that different types of substructure can be developed in tungsten single crystals deformed by rolling at temperatures in excess of 700°C. For the same percent reduction in thickness, the degree of work hardening is influenced by the slip mode which depends on (1) crystal orientation relative to the rolls and (2) the selected rolling parameters such as contact angle and roll velocity. Simple slip on conjugate planes at the crystal surfaces, resulting in duplex slip in the center of the crystal, represents the least work hardened condition of the rolled sheet. Multiple slip, on the other hand, results in more complex substructural interactions and consequently, a more work hardened crystal. Two of the selected combinations of rolling plane and direction showed very little change in hardness from 10% on the initial pass, to 50% reduction in thickness after several passes. Therefore the recovery studies were conducted on these two orientations, the (110) [001] and (001) [110]. The hardness was recorded after one-half hour anneals at various temperatures and correspondingly etch pit and Laue back-reflection photographs were taken to observe substructural changes.

Inasmuch as some of the results of the last year's program, especially on specimen deformed a small percentage, showed an extreme sensitivity to the surface conditions with regard to recrystallization, extra precautions were taken in this investigation. After rolling the surfaces were cleaned up by grinding and electrolytic polishing to remove any residual work due to grinding. This totalled to about a 10 mil reduction in thickness prior to the annealing treatments.

4.2.1 EFFECT OF TEMPERATURE

In this phase of the present program, the crystals were not given an initial one-half hour anneal at 2400°C prior to rolling. In last year's program, it was shown that annealing the as-grown crystal can reduce the dislocation density and the hardness; therefore after the initial deformation of 10%, a slightly higher hardness value was recorded for this series of crystals than those given a prior anneal.

After 10% deformation on the initial pass, the (110) [001] and (001) [110] crystals, show a decrease in hardness with increasing annealing temperature (Figure 13). The initial hardening was anisotropic and the direction with the higher hardness was found to recover more rapidly than the direction with the initially lower hardness. Approaching temperatures near 2400 or 2500°C, both the hardness in the horizontal direction of the transverse plane and the hardness in the vertical direction of the transverse plane begin to converge. They approach the hardness values of the as-grown crystals with the remaining difference due to the inherent anisotropy of hardness measurements in single crystals. (see Section 4.1.2).

After a one-half hour anneal at 2500°C, the (110) [001] crystal had a hardness value which was in excess of the values reported for the as-grown crystal give a 2400°C anneal prior to rolling. Since good furnace control could only be obtained at a maximum temperature of 2500°C, the recovery treatment involved increasing the time at 2500°C beyond one-half hour in order to see if the hardness values could be further decreased to those of the as-grown plus 2400°C annealed specimens. Prior to reporting these results, it should be reiterated that in all the annealing studies on crystals deformed 10%, absolutely no recrystallization was detected.

In last year's program, recrystallization was observed in specimens deformed 10%. These apparently conflicting results illustrate the sensitivity of recrystallization to the surface conditions; for in the previous study, no surface removal was done prior to annealing. Therefore, the high dislocation density present at the surface due to the high frictional stress between the rolls and the crystal during rolling can strongly influence the recrystallization tendencies of the deformed crystals - especially at low percent reductions.

4.2.2 EFFECT OF TIME AT TEMPERATURE

Increasing the time at 2500°C from one-half hour to three hours showed that the hardness could be further decreased to values approaching 330 BHN, which is the value for the as-grown annealed (110) [001] crystal.

In Figure 14, a series of magnified Laue asterisms are presented in order to illustrate the fragmentation of a Laue spot with time at temperature. In Figure 15, the accompanying change in dislocation density with annealing time at temperature is also illustrated with a series of photographs.

The observations can be summarized as follows:

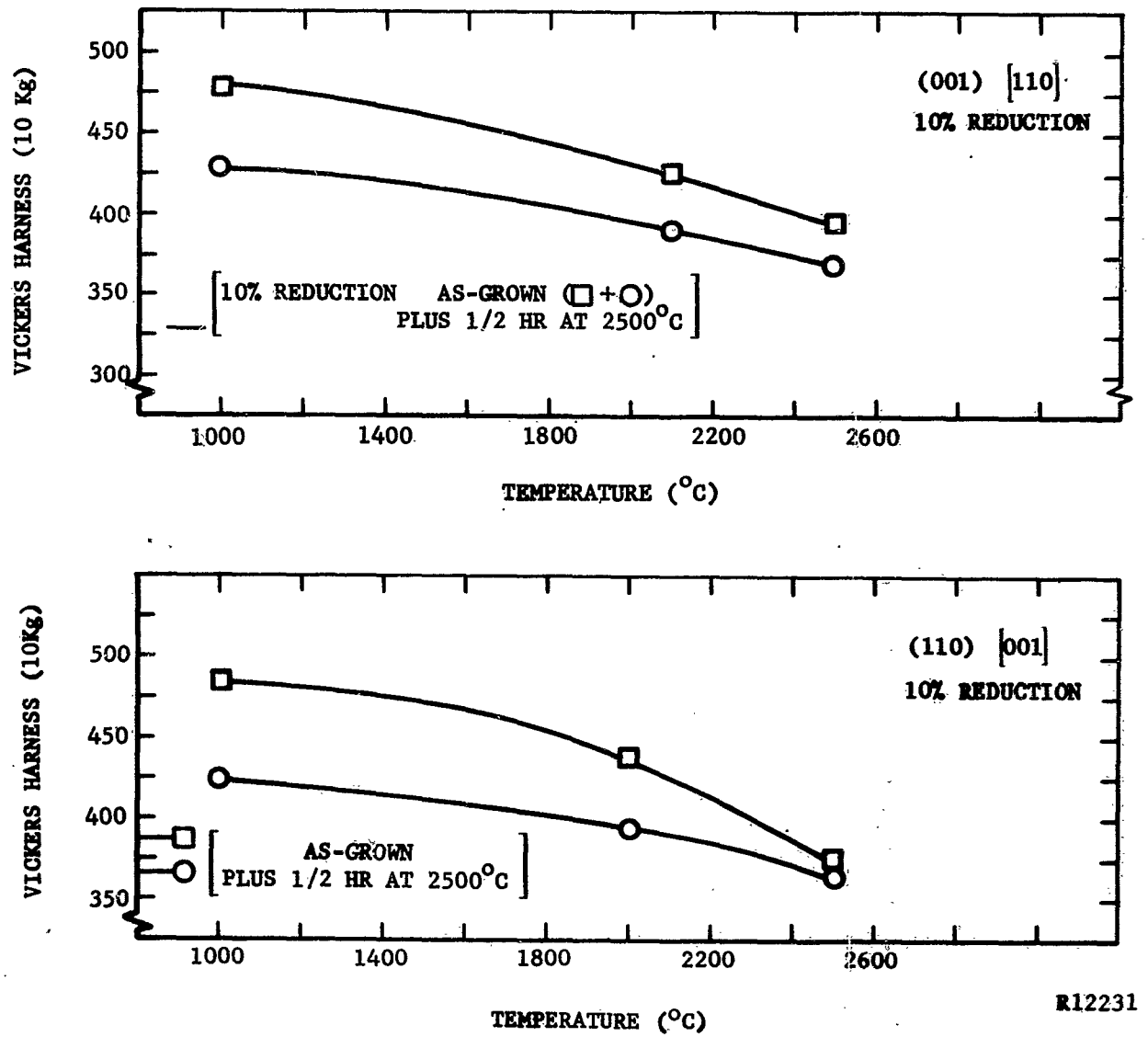
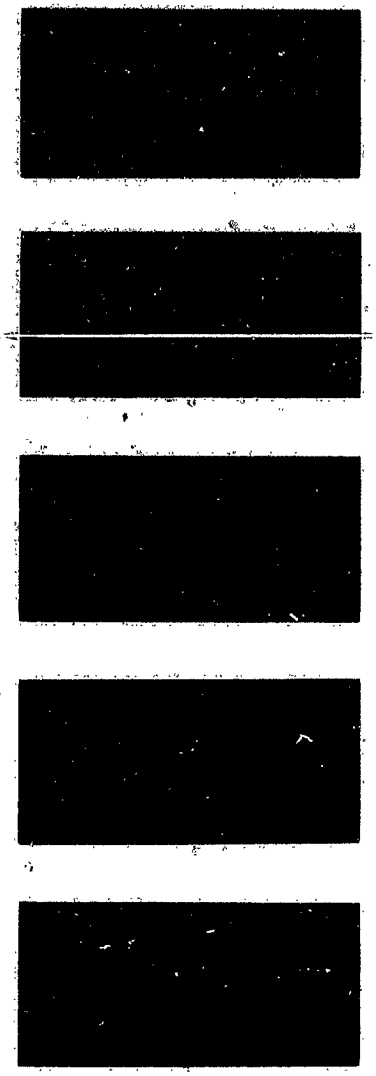


FIGURE 13. THE EFFECT OF A ONE-HALF HOUR ANNEAL AT VARIOUS TEMPERATURES ON THE HARDNESS AFTER A 10 PERCENT REDUCTION IN THICKNESS BY ROLLING. NO RECRYSTALLIZATION OCCURRED AT INDICATED TEMPERATURES.



(A) AS-ROLLED

(B) 1/2 HR - 2500°C

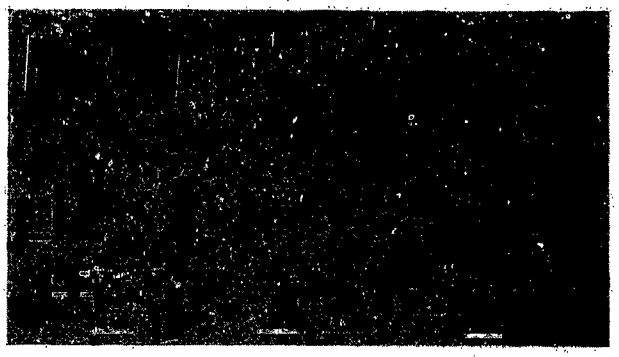
(C) 2-1/2 HR - 2500°C

(D) 3-1/2 HR - 2500°C

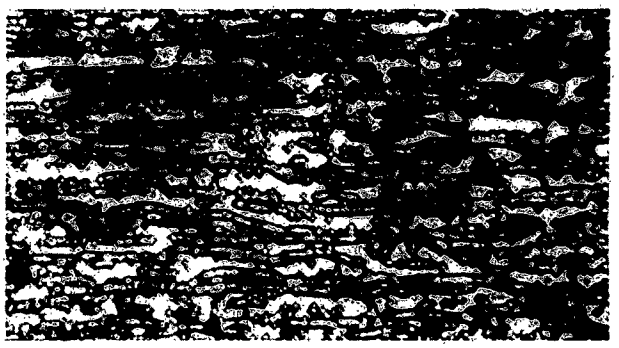
(E) 3-1/2 HR - 2500°C
PLUS 1/2 HR - 2800°C

R12232

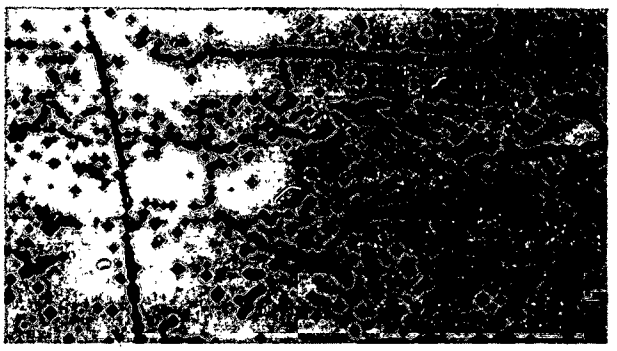
URE 14. FRAGMENTATION OF LAUE SPOTS ON ANNEALING (100) [011] CRYSTAL REDUCED 10 PERCENT IN THICKNESS. NO RECRYSTALLIZATION WAS OBSERVED.



(A) AS-GROWN (250X)



(B) 10 PERCENT REDUCTION (500X)



(C) 2-1/2 HRS AT 2500°C (250X)

R12233

FIGURE 15. RECOVERY OF THE DISLOCATION DENSITY ON ANNEALING THE (100) [011] CRYSTAL REDUCED 10 PERCENT IN THICKNESS. NO RECRYSTALLIZATION WAS OBSERVED.

(1) With deformation, the as-grown dislocation density (Fig. 15A) is increased from about 10^5 per cm^2 to 5×10^7 per cm^2 with 10% reduction. (Fig. 15B). Correspondingly, lattice bending is observed in one direction (Fig. 14A).

(2) A one-half hour anneal at 2500°C causes an abrupt decrease in dislocation density to about 10^6 which changes very little with time at temperature (Fig. 15C). Correspondingly, fragmentation of the Laue spot becomes more pronounced with time at temperature (Figs. 14 B, C and D). This fragmentation is related to the formation of subgrain boundaries (Fig. 15C).

(3) An additional one-half hour anneal at 2800°C increases the fragmentation of the Laue spot (Fig. 14E) which is accompanied by a greater density of subgrain boundaries. (Compare Figs. 16A and 16B).

Therefore after 3½ hours at 2500°C and ½ hour at 2800°C, residual lattice bending is retained although the dislocation density and hardness are approaching values found for the as-grown crystals.

Another interesting sequence of pictures taken on this crystal, illustrates the coalescence of dislocation as they annihilate during the recovery treatment at 2500°C (see Fig. 17). It appears as a spider web (Fig. 17A). At low magnifications, the core of the web gives the appearance of a new grain; but at magnifications of 1000x, using dark field illumination (Fig. 17B), the core is seen to be nothing more than an extremely high density of dislocations. That no change in lattice orientation exists within the core is verified by comparing the shape of the etch pits within the core, to the shape of the pits in the surrounding area. Therefore, the core is but a sink for dislocation annihilation during the recovery process.

4.2.3 EFFECT ON PROPERTIES

Since 10% reduction in thickness resulted in very little work hardening for the (110) [001] and (001)[110] crystals, annealing treatments caused very little detectable changes in the shapes of the stress-strain curves (Figs. 10 and 11). But it could be generalized that, as the annealing temperature was increased and complete recovery was approached, the stress-strain curves approached that given by the annealed crystals.

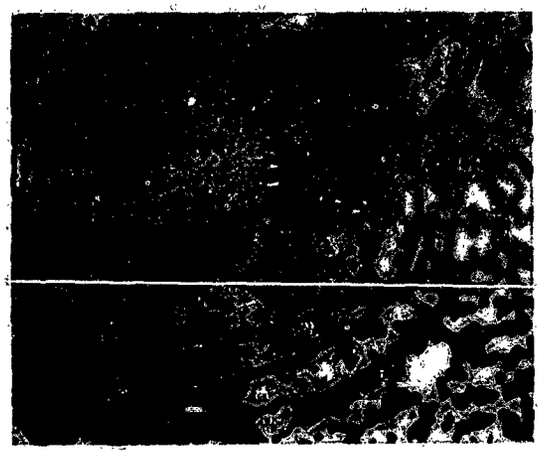
Specimens reduced 50% in thickness by multiple-pass rolling techniques were given a one-half hour anneal at 1200°C. This value was selected because the 1000°C anneal caused no change in hardness and no change

(A) 1-1/2 HR AT 2500°C (250X)

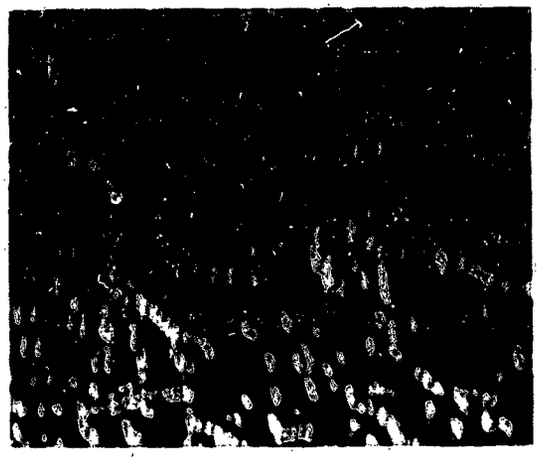
(B) 3-1/2 HR AT 2500°C (250X)
PLUS 1/2 HR AT 2800°C

R12234

FIGURE 16. SAME AREAS ILLUSTRATING THE INCREASE IN DENSITY OF SUBRAIN BOUNDARIES ON ANNEALING



(A) (385X)



(B) (1000X)

R12235

FIGURE 17. SPIDER WEB EFFECT AS DISLOCATIONS ANNIHILATE DURING RECOVERY TREATMENT AT 2500°C

in dislocation density or Laue back-reflection photographs; and above 1400°C, recrystallization began to take place (see Section 4.3).

The most significant effect of a 1200°C annealing treatment i.e., stress relieving treatment, was the tendency to cause embrittlement. This tendency was most pronounced in crystals deformed by multiple slip. Crystals deformed under conditions of simple and duplex slip, showed little or no tendency to embrittle. Since no gross changes in dislocation substructure are detected after ½ hour at 1200°C, it appears that the embrittling effect must be due to the formation of interstitial atmospheres around dislocations, even in the ppm range. Also, since the effect is most pronounced in crystals deformed by multiple-slip, there must be an enhanced dislocation-atom interaction due to the greater misfit at points of intersection of dislocations. That is, at the node of intersecting dislocations the tendency must be greater to form Cottrell atmospheres than along the length of a dislocation line.

4.3 RECRYSTALLIZATION

In last year's program, the recrystallization studies showed a difference in recrystallization temperature of about 200°C due to the difference in crystal orientation. Now that the deformation characteristics of rolled single crystals have been rigorously defined with the first phase of this year's program, some of the observations on recrystallization can now be understood.

For the same percent reduction in thickness, four different work hardened conditions can exist. These are:

(1) Easy glide or simple slip found at the surfaces of the crystal,

(2) Duplex slip occurring in the center of the crystal. Conditions (1) and (2) have been observed in the (001) [110] and (110) [001] orientations.

(3) Multiple slip. Although there are degrees of the severity of this type of deformation, in general it represents a more complex deformation mode than (1) and (2). Also the deformation is rather uniform throughout the crystal.

(4) High surface density of dislocations due to the frictional shear stress between the roles and the crystal. This effect is present in all orientations.

As in the recovery studies (see Section 4.2), the surface effects can mask the behavior of the other less work-hardened states of deformation such as easy glide and duplex slip. Therefore, the crystals were ground to clean up the surface and polished to remove the effects of grinding. The entire operation involved a total removal of 10 mils from the thickness. Again the (110) [001] and (001) [110] crystals were used. The specimens were reduced by 50% in thickness by multiple pass rolling techniques so that the least work hardened condition of the crystal existed. In fact, the hardness (Fig. 4) does not change significantly above that obtained after 10% reduction in thickness. It was felt that the most severe conditions of deformation were well represented and analyzed in last year's program.

4.3.1 REMOVAL OF HARDNESS

For the two orientations, 50% reduction in thickness directionally increases the hardness. A one-half hour 1000°C anneal does not change the hardness; but at higher temperatures the hardness continues to decrease - in fact, at a rate similar to the recovery rate for the crystal reduced 10% (see Fig. 13). At a temperature between 1400°C and 1600°C, a precipitous drop in hardness occurs (Figs. 18 and 20). The temperature at which this drop in hardness occurs is independent of crystal orientation. In fact, it is identical to that recorded on heavily worked powder metallurgy tungsten[5] and reported in last year's program[6].

But in the previous program,[6] a 200°C difference in recrystallization temperature was recorded. Obviously then, the characteristic curve for the removal of hardness with annealing does not accurately define the degree of recrystallization. In fact, it will be presently shown that for the proper combination of deformation and crystal orientation, the recrystallization process can be a two step process i.e., (1) the complete removal of hardness without reorientation and (2) the appearance of differently oriented grains.

4.3.2 FORMATION OF GRAIN BOUNDARIES

Figure 18 is the annealing curve for the (110) [001] crystal reduced 50% in thickness by multiple-pass rolling techniques. Superimposed is the plot of percent recrystallization versus annealing temperature. Upon close examination of this curve, some very interesting and technically significant issues were generated.

A discrepancy occurred between the hardness taken on the transverse plane and the hardness taken on the rolling plane after a 1600°C anneal. Correlation with microscopic observation showed a resulting sandwich structure which is illustrated in Figure 19. The center of

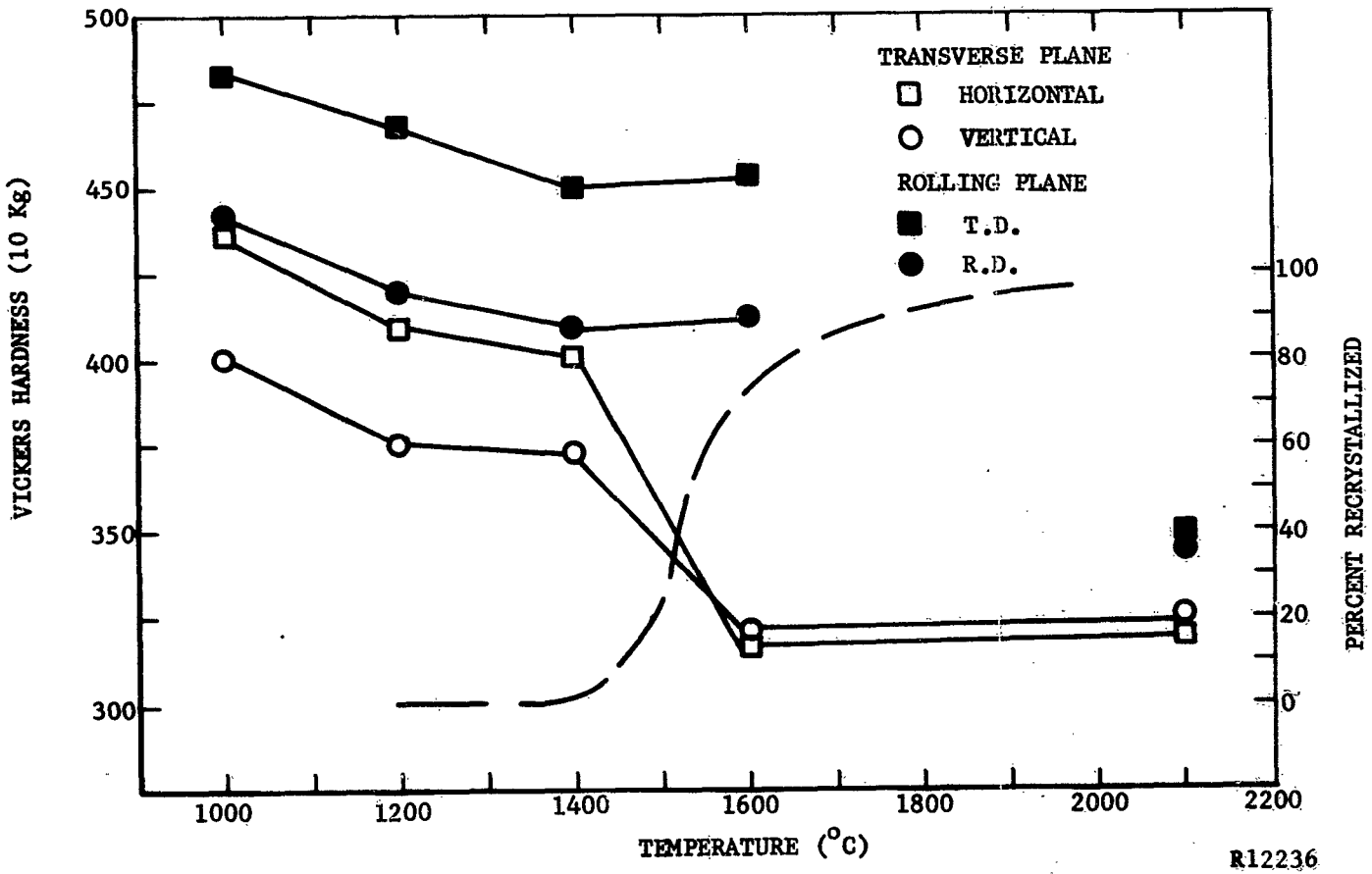


FIGURE 18. ANNEALING CURVE OF (110) [001] CRYSTAL REDUCED 50 PERCENT IN THICKNESS IN SIX ROLLING PASSES

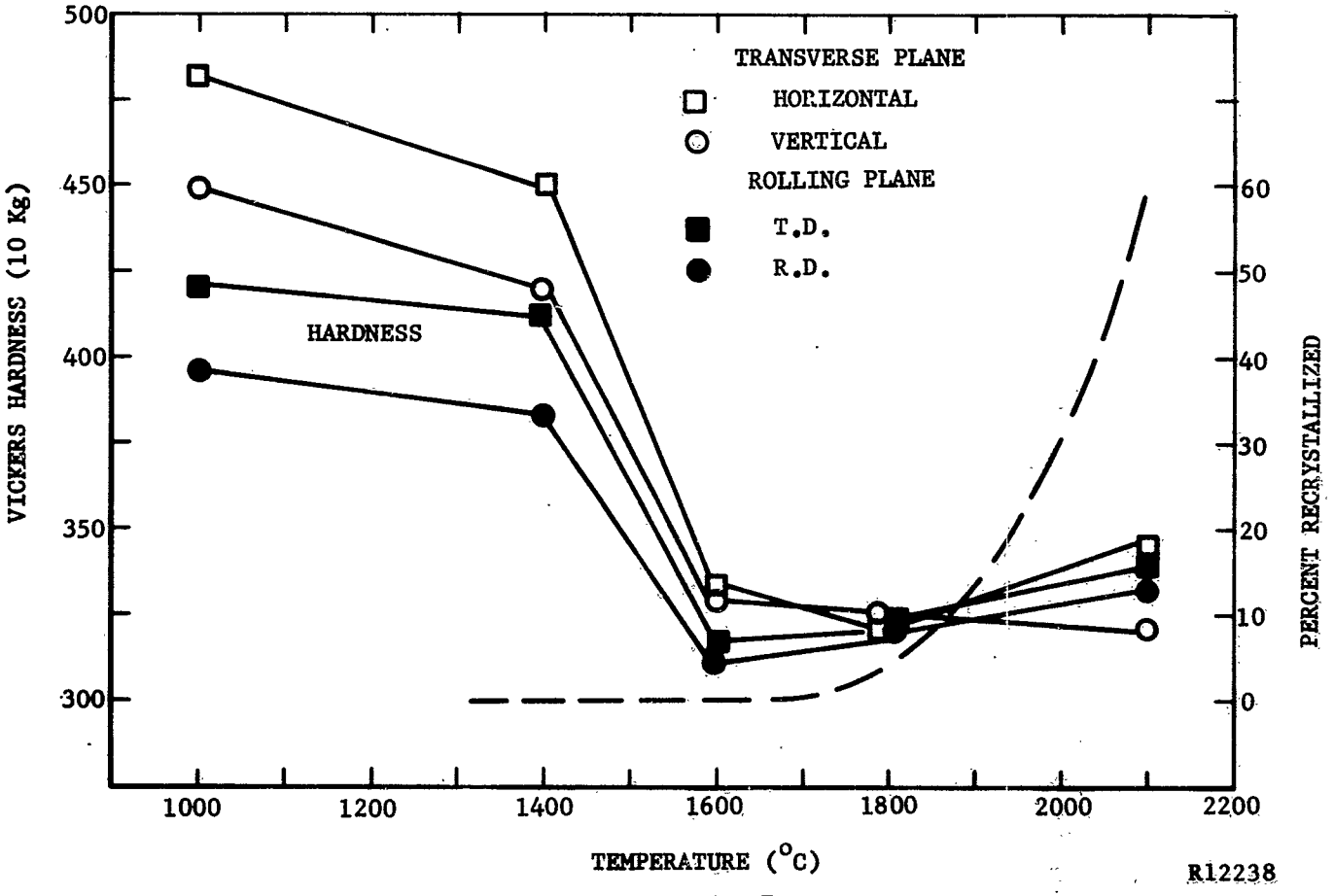


FIGURE 20. ANNEALING CURVE OF (001) [110] CRYSTAL REDUCED 50 PERCENT IN THICKNESS IN SIX ROLLING PASSES

the thickness section was recrystallized whereas the surface was not. If we refer to the deformation characteristics of this orientation, it is realized that simple slip occurred at the surface and duplex in the center. The hardness after a one-half hour 1600°C anneal remained the same at the surface, that is, it followed the typical annealing curve for a crystal undergoing recovery as observed in Fig. 13. The hardness values taken in the center of the transverse plane had hardness values correlating to a completely recrystallized grain. The crystal was about 60 to 70% recrystallized. The issue is: regions deformed by duplex slip tend to recrystallize before regions deformed by easy glide. Therefore, the region of duplex slip represents a more work hardened region than the regions deformed by simple slip.

Since the deformation characteristics are the same for the crystal (100) [110], which is also the recrystallization texture of polycrystalline sheet materials, the same recrystallization behavior would be expected. If Figure 20 is examined, a precipitous drop in hardness is again observed at temperature near 1600°C; but no recrystallization has occurred. In other words, <u>complete softening has preceded the onset of recrystallization!</u> Grain boundaries are finally observed for one-half hour anneals at temperatures of 2100°C. This same behavior has been previously reported in a single crystal of aluminum which was also deformed with its rolling orientation the same as the recrystallization texture.[7] It is essential that this rolling orientation be retained throughout deformation. Similar requirements exist in our investigation.

How does the substructure differ when abrupt softening occurs at 1600°C as compared to long times at temperatures in excess of 2500°C? At best it can be stated that (1) the dislocation density is slightly higher at 1600°C than 2500°C, (2) subboundary formation is less distinct at 1600°C than 2500°C and (3) residual lattice bending is absent at 1600°C in comparison to its persistance at 2500°C. The hardness values are similar; a higher dislocation density existed originally in the specimen worked 50% as compared to 10%; and the lattice bending was originally greater in the 50% than after the 10% reduction in thickness.

Since the decrease in hardness with annealing temperature was similar for the 10% and 50% deformed crystals, the decrease in density of the dislocation substructure attending this process is attributed to recovery. If this recovery rate were extrapolated to 1600°C, as fortuitously evidenced at the surface of the (110) [001] crystal (Fig. 18), the observed drop in hardness also observed at 1600°C must be relatively described as precipitous and the associated softening process given a designation other than "recovery". Therefore, the process involving complete softening without the onset of recrystallization must be differentiated from the recovery process and therefore aptly be described as recrystallization "<u>in situ</u>".[6]

4.3.3 EFFECT ON PROPERTIES

It has been illustrated that the introduction of grain boundaries does not significantly alter the strength of tungsten[8] but it does catastrophically affect the ductility.[8] Since another source of data has made this comparison, it will be withheld until the discussion (Section 5). Although several of our single crystal tensile specimens were recrystallized and tested at room temperature in order to confirm the embrittling effect of grain boundaries, no further quantative data was generated at elevated temperatures since our major concern is obtaining room temperature ductility.

4.4 METALLURGICAL PROPERTIES

4.4.1 FABRICABILITY

Polycrystalline products are fabricated by initially forging, rolling, or swaging of the sintered compacts at 2900°F to 3300°F (1600°C to 1820°C); further working is done at successively lower temperatures as the reduction in area proceeds.[9] Single crystals, on the other hand, are grown by the Arc-Verneuil technique.

The Arc-Verneuil technique is an adaptation of the flame-Verneuil crystal-growth method which has been operated for many years to grow single crystals of sapphire, ruby, rutile and other oxides which melt at temperatures below about 2200°C. In its most simple form the process consists of a flame which impinges upon a vertically-held seed rod within a furnace. The molten cap produced at the top of the rod creates a solid-liquid interface without the need for auxiliary contaminating crucibles or refractory materials. Fine powder of the material is fed through the flame and drops onto the molten cap. The seed rod is lowered through the furnace at a slow, uniform rate. Under steady-state conditions the solid-liquid interface, which represents a plane of constant temperature at the crystal melting point, remains positionally fixed in the furnace. Except for powder addition which fails to impinge on the cap, the rate at which the crystal is withdrawn from the hot zone is equal to the rate of powder addition.

The flame process has two principal limitations: only materials with melting points below the flame temperature can be grown; and the crystal must be chemically compatible with the combustion products of the hydrogen-oxygen (or other high temperature) reaction. An inert atmosphere plasma frees the process from these limitations. An arc device can be substituted for the combustion burner so that a hot inert-atmosphere effluent impinges upon the seed crystal. This electrical analogue of the falme approach retains the main advantage of the Verneuil method in that the hot growing crystal does not come in

contact with a reactive container. Surface tension maintains the molten cap perched upon the top of the seed. The arc heat source permits growth of high-melting-point reactive materials such as refractory metals, carbides, borides and oxides. A self-explanatory, schematic drawing of the equipment used for the growth of tungsten crystals is shown in Figure 21.

The process can be scaled-up to allow the preparation of very large crystal sizes. Cylindrical-shaped boules have been grown with diameters ranging from 1/4 inch to over one inch with lengths greater than 15 inches (Figure 22). Slab-shaped boules, preferable for subsequent rolling, can also be grown to sizes of 1½ x 3/8 x 12 inches (Figure 23). Any desired rolling plane and rolling direction can be obtained by using preoriented seeds. Solid solution alloying is also possible. The initial forging, rolling or swaging temperature of the single crystal billet is about one-half that of sintered products, i.e., 1500°F to 1800°F (800°C to 1000°C).

4.4.2 ROLLABILITY

The ability of the crystal to deform to the desired roll setting, which might be termed "rollability", is dependent on the crystal orientation relative to the rolls. It can be again explained on the basis of the number of operating slip systems. Deformation by a combination of easy glide and duplex slip, which is found in the (011) [110] and (110) [001], has the lowest degree of work hardening. Because of the self-alligning nature of the two slip systems in the (001) [110] (see Section 5.1), it is the easiest orientation to work with and obtain desired settings. Obviously the (110) [001] is second, followed by the (100) [001] and then the (110) [110], which has a maximum number of slip systems operation and thus work hardens most rapidly.

4.4.3 WELDABILITY

Welding tungsten to itself by most conventional welding procedures results in an embrittled joint. Figure 24 shows the cross-sectional view of electron beam welded polycrystalline and single crystal tungsten. In contrast to the typical weld joint of a recrystallized and heat affected zone, the single crystal is seen to be absent of any recrystallization. No effects of the weld can be observed. Only by x-ray can slight lattice misorientations be detected in the weld zone. Therefore, any embrittling effects due to welding would be absent in joining single crystals.

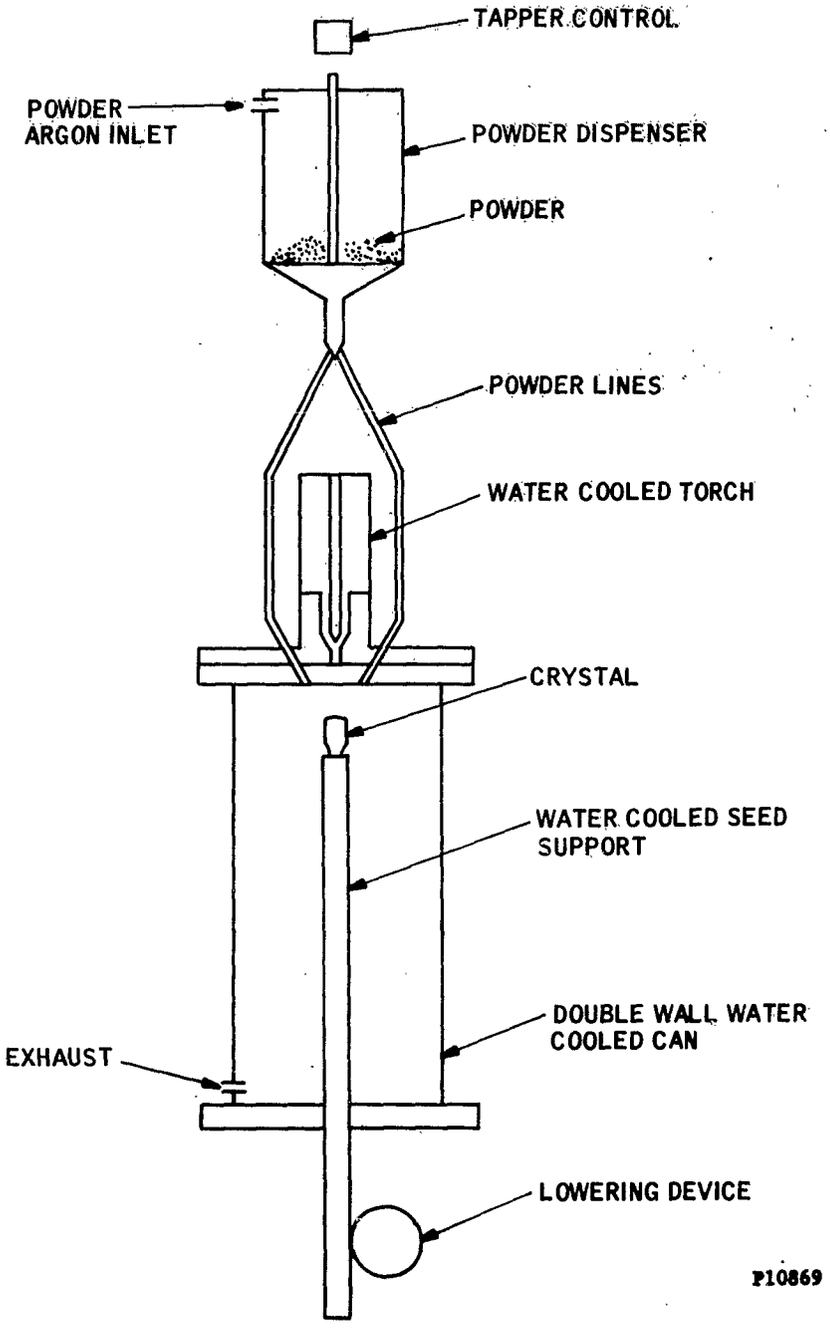


FIGURE 21. EQUIPMENT FOR GROWING SINGLE CRYSTALS

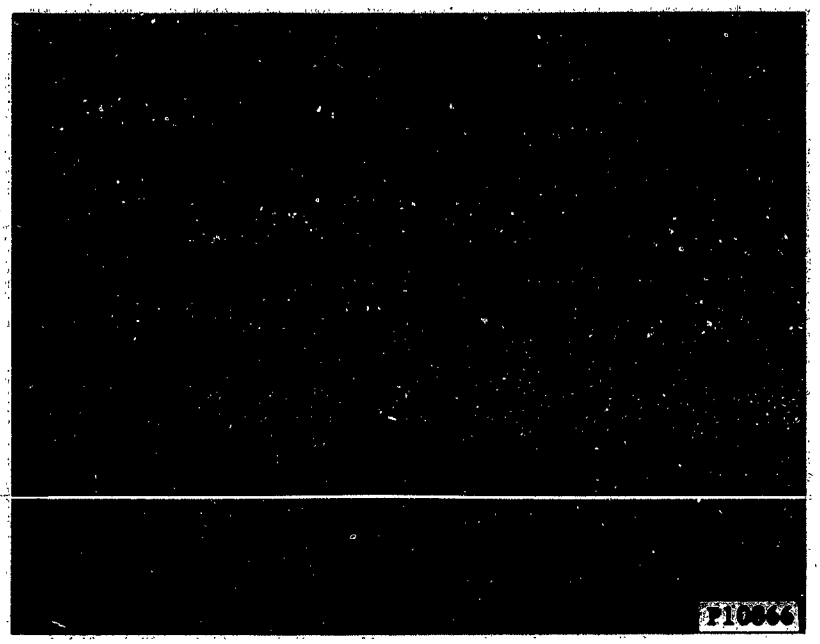
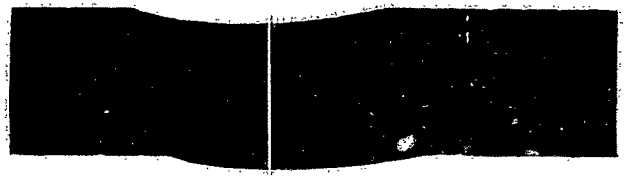


FIGURE 22. CYLINDRICAL-SHAPED BOULES

FIGURE 23. SLAB-SHAPED BOULE

SINGLE CRYSTAL

POLYCRYSTALLINE



NO RECRYSTALLIZATION IN WELD ZONE

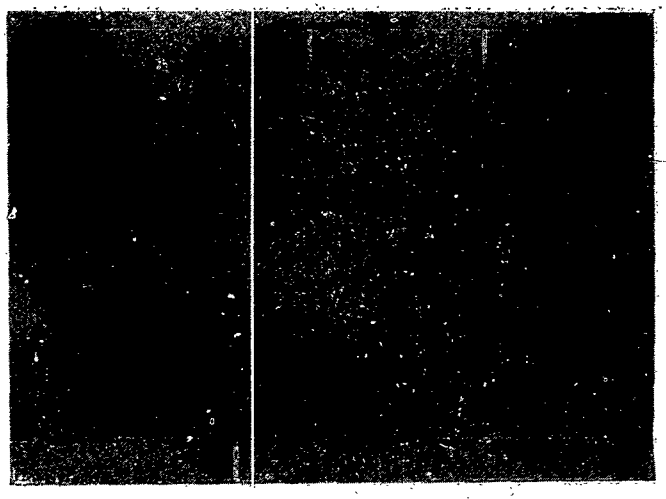
TYPICAL RECRYSTALLIZED WELD ZONE

BUTT WELD

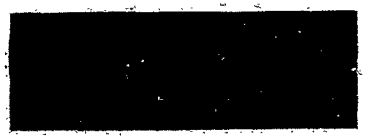
a. NO PREHEAT

b. NO CRACKS

c. NO RECRYSTALLIZATION IN WELD ZONE



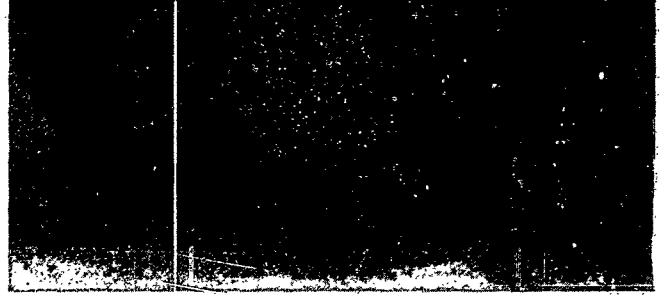
1. BENT 1200°F
2. WELDED
   a. NO PREHEAT
   b. NO DISTORTION



a. NO PREHEAT

b. NO CRACKS

c. NO RECRYSTALLIZATION IN WELD ZONE

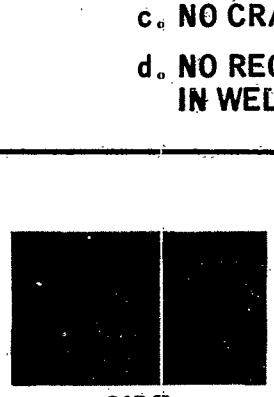


1. BENT 1200°F
2. WELDED
   a. NO PREHEAT
   b. NO DISTORTION
   c. NO CRACKS
   d. NO RECRYSTALLIZATION IN WELD ZONE

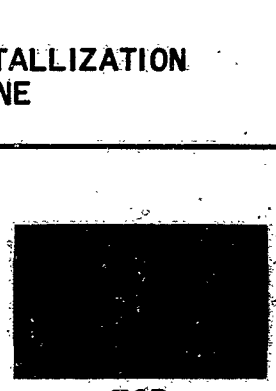
TOP

SIDE

1. STRAIGHT SECTION WITH THREE E.B. WELDS
   a. NO PREHEAT
   b. NO CRACKS
   c. NO RECRYSTALLIZATION IN WELD ZONE
2. BENT TO PRESENT SHAPE AT R.T.



SIDE



TOP

1. BENT TO 45° AT -100°F
2. E. B. WELDED
   a. NO PREHEAT
   b. NO DISTORTION
   c. NO CRACKS
   d. NO RECRYSTALLIZATION IN WELD ZONE
3. BENT FURTHER TO 90° AT R.T.

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FIGURE 24. FORMING AND JOINING SINGLE CRYSTAL SHEET USING ELECTRON BEAM WELDING (APPROX. 2X)

Also illustrated in Figure 24 are different sequences of forming and joining by Electron Beam (EB) welding. Crystals initially deformed as low as -100°F, can be welded without recrystallization occurring in the weld zone and therefore, be further deformed at room temperature without the initiation of brittle fracture. (see Fig. 24). In all welds no distortion was detected and no preheat was necessary prior to welding. Under these conditions no cracks were detected, further illustrating the resistance of single crystals to thermal shock.

Polycrystalline tungsten sheet, by contrast, needed preheat, had large grains in the weld zone, and the weld became a source of brittle fracture.

4.4.4 BEND TRANSITION TEMPERATURE

Since one prime object of this program was to retain the tungsten in an un-worked monocrystalline condition, the ductile-to-brittle transition temperature was evaluated for the as-grown crystals. The evaluation was in conformity with the MAB test procedures.[10] A punch radius of four times the thickness of the sheet, which was 0.025 inches. The remaining dimensions were 0.25 x 1.25 inches with the surfaces previously ground and polished (10 mils removed) prior to testing. The crosshead speed was 1.0 inch per minute and the specimens were bent to a free bend angle of 90°. The transition temperature was defined as the lowest temperature to which a sound 90° bend can be produced for a 4t radius punch. The results are listed in Table V for four different combinations of rolling plane and rolling direction.

TABLE V

BEND TRANSITION TEMPERATURE OF ANNEALED TUNGSTEN SINGLE CRYSTALS (8t)

Orientation	DBTT
(110) [$\bar{1}$10]	-50°F
(001) [100]	-60°F
(001) [110]	-75°F
(001) [110]	-125°F

Although the punch radius was 4t, the radius of curvature of the first three orientations resulted in an 8t bend. The obvious reason for this inconsistency is the inherent anisotropy of the single crystal. The (001) [110] on the other hand, conformed to the prescribed 4t punch radius -- and at this radius, had a DBTT at minus 75°F. Increasing the punch die radius to 8t in order to compare the results to the other

three orientations, lowered the DBTT (as listed in Table V) to minus 125°F.

In any future bend test evaluations, it would be more practical to obtain the minimum bend radius at room temperature by measuring the true bend radius. This radius need not correlate to the punch die radius. The anisotropic behavior of the sheet should be accounted for in die design for the eventual utilization of this product. It should be further pointed out that in all bend tests, or for that matter, in any of the machining or grinding operations in preparing specimens for this program, delamination was never observed.

SECTION 5

DISCUSSION

5.1 ON DEFORMATION BY ROLLING

While no attempt was made to clarify the deformation processes occurring in the (110) [$\bar{1}$10] and the (001) [100] crystals, the deformation mode of the remaining two crystal orientations is very clear. The observations of the macroscopic dimensional changes, x-ray photographs, hardness, and dislocation etch pits can all be explained by the fact that slip has occurred on the two conjugate {112} <111> systems having the highest resolved shear stress.

For small reductions, i.e. for zero contact angle, the stresses in rolling can be considered to consist of a compressive stress σ_c normal to the rolling plane and tensile stress σ_t in the rolling direction. The shear stress acting on a particular slip plane and in a particular slip direction can be calculated from the relationship:

$$\tau = \sigma_c \cos \phi_c \cdot \cos \lambda_c + \sigma_t \cos \phi_t \cdot \cos \lambda_t \qquad \text{Eqn. (1)}$$

where ϕ_c = angle between the compressive stress direction and the normal to the slip plane.

ϕ_t = angle between the tensile stress direction and the normal to the slip plane.

λ_c = angle between the compressive stress direction and the slip direction.

λ_t = angle between the tensile stress direction and the slip direction.

In order to illustrate the {112} <111> slip systems that have the highest resolved shear stress component of σ_c, the ideal rolling orientation of the four crystals is shown in Figure 25. The influence of superimposing the shear stress component of σ_t will be discussed separately for each crystal orientation.

5.1.1 (001) [110] ORIENTATION

For the (001) [110] crystal, four slip systems have the same maximum resolved shear stress due to the compressive stress i.e., $\tau = 0.47\ \sigma_c$; but two of them, lying in the rolling direction, have an additional shear stress due to the tensile stress i.e., $\tau = 0.47\ \sigma_t$. For this reason, only the latter two slip systems would be expected to operate, resulting in duplex slip.

For these two systems, the slip direction is at an angle of 35° to the rolling direction. If by slight misorientation either from growth or during rolling the resolved shear stress is higher on one of the two slip systems, only this one system will be active. The resulting rotation of this plane towards the rolling direction will automatically cause the conjugate system to become more favorable i.e., the maximum value of the resolved shear stress shifts to the conjugate system.

The dislocation etch pit studies proved that only these two conjugate {112} planes were indeed the operative slip planes, and the Laue photographs identified the slip directions as the <111> direction, lying in these planes. The absence of billet widening and the small amount of hardening in the vertical direction are in complete agreement with this deformation model.

5.1.2 (110) [001] ORIENTATION

The (110) [001] crystal has two slip systems, both lying in the rolling direction, with a maximum resolved shear stress due to the combined tensile and compressive stresses again given by $\tau = 0.47\ (\sigma_t + \sigma_c)$. These two slip systems are symmetrical about the rolling plane with the direction of the rolling stress, direction of slip, and normal to the slip plane all coplanar. For perfect crystal orientation relative to the rolls, deformation should ideally occur by duplex slip, resulting in no change of the original crystal orientation and elongation only in the rolling direction. Again, as in the case of the (001) [110] crystal, the experimentally determined slip planes and directions agreed with the ones shown in Fig. 25.

While the behavior and results of the (001) [110] and the (110) [001] crystals seem to be the same, there is however one essential difference: in the (110) [001] crystal the slip directions are inclined at an angle

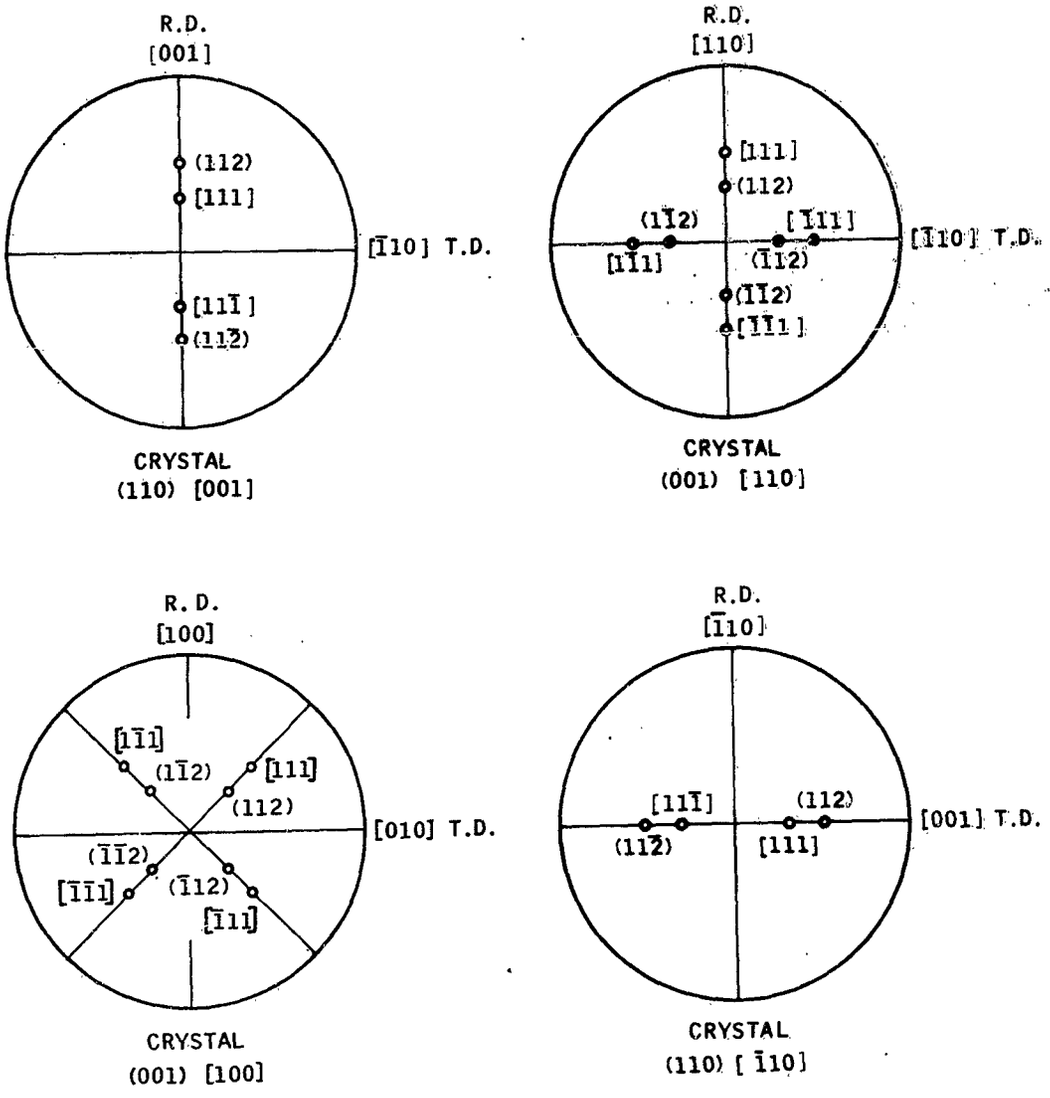

FIGURE 25. SLIP SYSTEMS WITH HIGHEST RESOLVED SHEAR STRESS UNDER A COMPRESSIVE STRESS NORMAL TO ROLLING PLANE AND TENSILE STRESS IN ROLLING DIRECTION: o-ACTIVE SYSTEMS, ●-INACTIVE SYSTEMS

of 55° to the rolling direction. If by slight misorientation one of the two slip systems has a slightly higher resolved shear stress and only one system initially becomes active, the resolved shear stress for the active system will continuously increase with rotation of the lattice, in contrast to the self-stabilizing behavior of the (001) [110] crystal. The imperfect duplex slip of the (110) [001] crystal manifested itself by a rolling curvature in the crystal and can only be corrected by modifying the rolling procedure, as experienced in the rolling program.

Polanyi[11] has already pointed out, that it is the stability of the (001) [110] orientation that is the explanation for the (001) [110] being the rolling texture of polycrystalline tungsten sheet.

Once the slip systems have been established, the occurrence of simple slip instead of duplex slip close to the rolling surfaces (Fig. 9) can now be explained by the presence of a finite contact angle. For the upper surface, for example, the tensile stress is tilted downward, thereby increasing the resolved shear stress for one system and decreasing it for the conjugate system. Since the slip directions are inclined at 55° against the rolling direction for the (110) [001] crystals as compared to 35° for the (001) [110] crystal, the effect is the opposite for the two billets.

The contact angle or reduction per pass has also an important influence on the deformation mode as indicated in Table 4. For the (110) [001] crystal duplex slip was observed up to 20% thickness reduction in one pass, although the lattice bending increases with increasing contact angle. However for a 12° contact angle (53% reduction), multiple slip has occurred. Therefore it appears that a critical contact angle exists, beyond which deformation will not occur by duplex slip. Whether the concomitant increase in strain rate influences the deformation mode, has yet to be determined.

5.1.3 (001) [100] ORIENTATION

The (001) [100] crystal has an entirely different deformation behavior. With the same rolling plane as (001) [110] crystal, but the rolling direction rotated 45°, all four systems now have the same maximum resolved shear stress, $\tau = 0.47\,\sigma_c + 0.23\,\sigma_t$. As a result, slip occurs on two pairs of conjugate slip systems belonging to two different zones. Dimensional changes, Laue photographs and the high isotropic hardening confirm this duplex deformation mode.

5.1.4 (110) [$\bar{1}$10] ORIENTATION

A complex deformation similar to the (001) [100] crystal is also observed for the (110) [$\bar{1}$10] orientation. This crystal is only different

from the (110) [001] crystal, in that the rolling and transverse directions are exchanged. The two most favorable {112} <111> slip systems lie in the transverse direction, with a resolved shear stress $\tau = 0.47\ \sigma_c$. The tensile shear stress component contributes nothing to these two most favorable slip systems. On the second and subsequent passes several other slip systems were operative; but it seems that during the first pass, only the above mentioned two slip systems were active since broadening but no elongation of the billets was observed.

5.1.5 ROLLING PARAMETERS

In order to predict deformation characteristics of single crystals during rolling, the ratio of tensile stress σ_t to the compressive stress σ_c, as well as the strain rate are of considerable interest. The tensile or frictional stress is related to the compressive stress by the equation: $\sigma_t = \mu \cdot \sigma_c$, where μ is the coefficient of friction. For hot rolling a typical value of μ is about 0.5. An estimate of μ in the present work can be made by comparison of the dimensional changes of two billets. During the first pass, the same pair of conjugate slip systems were active for both the (110) [001] and the (110) [$\bar{1}$10] crystal, but the behavior of the two billets during rolling was quite different. From Table I it can be seen, that for constant roll-setting and the same original thickness of both billets, the (110) [001] crystal showed twice as much plastic strain as the (110) [$\bar{1}$10] crystal:

$$(110)\ [001] \quad : \quad \epsilon_w = 0.00,\ \epsilon_l = 0.14$$
$$(110)\ [\bar{1}10] \quad : \quad \epsilon_w = 0.07,\ \epsilon_l = 0.00$$

The larger strain of the (1$\bar{1}$0) [001] crystal is undoubtedly due to the higher resolved shear stress.

In order to determine μ, the roll-pressure as well as the stress-strain curve for this crystal orientation have to be known. In the absence of these data, an estimate of μ can be obtained by making the following simplifying assumptions:

1) The contact angle is zero.
2) The roll pressure is the same for both crystals.
3) The resolved shear stress is a linear function of the shear strain.

It follows that $\mu \approx 1$ and $\sigma_t \approx \sigma_c$.

For a calculation of the strain rates employed in the present work, the strain rate equation for sticking friction can be applied, since μ has been shown to have a high value. Therefore,

$$\bar{\dot{\epsilon}} = V[2/D(h_o - h_f)]^{1/2} \ln h_o/h_f \qquad \text{Eqn. (2)}$$

where

$\bar{\dot{\epsilon}}$ = mean strain rate

V = surface velocity of rolls

D = diameter of rolls

h_o = original thickness

h_f = final thickness

With billet thicknesses ranging from 0.25 inch to 0.1 inch, and passes of 10% reduction, the strain rates are about 200 to 400 $min.^{-1}$

The relationship of lattice bending to the starting billet thickness for a constant contact angle was also evaluated. Type (110) [001] crystals were used. A reduction of 25 mils with six inch diameter rolls corresponds to a contact angle of 5.2^o. With a 0.25 inch billet, the lattice bending is recorded at about 5^o. With a 0.129 inch billet reduced 26%, the contact angle is 6.1^o. The Laue photograph for the thinner crystal is now no longer unidirectional but instead, distorted in several directions as confirmed by 12% widening of the billet. With a yet thinner starting billet size of 0.077 inch which is then reduced 22%, the contact angle is but 4.2^o. Under the final conditions, Debye rings are apparent and 13.5% widening of the billet is recorded.

Since the contact angle was nearly constant, the variation in the mode of slip with decreasing specimen thickness can only be accounted for by either (1) a more severe quenching effect of the rolls on the thinner samples or (2) an increased strain rate. The strain rate dependence can be estimated from Equation 2.

Therefore the strain rate for the 0.25, 0.129, and 0.077 inch thick crystals is calculated as 4.2×10^3, 0.5×10^3 and 0.55×10^3 in./min., respectively. Since the last two values are not vastly different, the more rapid rate of cooling with decreasing specimen thickness must be responsible for inducing the secondary slip systems -- resulting in a more complex mode of slip. If accurate, this final result suggests the need for a higher roll temperature in order to maintain a simple mode of slip in thin sections. Under optimum conditions, one might expect the simple slip mode to be retained for reductions in excess of 50 or 60%.

5.2 ON RECOVERY AND RECRYSTALLIZATION

The substructural changes in tungsten single crystals deformed by rolling at temperatures in excess of 700°C can be explained on the basis of one slip system, the {112} <111>. The rolling procedures can be controlled to produce various types of deformation in the crystal. These modes of deformation encompass: (1) simple slip, (2) duplex slip, (3) multiple slip and (4) a high local dislocation density at the rolling surfaces due to the frictional stress. Each mode of deformation represents different degrees of work hardening within the crystal for a given percent reduction in thickness by rolling.

These different levels of work hardening produce different responses to annealing treatments. As anticipated, the recrystallization temperature is highest if deformation occurs by simple slip. Lower recrystallization temperatures are observed if deformation occurs by duplex slip and still lower for deformation by multiple slip. The local surface effects due to the frictional stress are the most difficult to define but they do represent a work hardened condition comparable to multiple slip. For example, the initiation of recrystallization can be changed from the surface to the center of the crystal sheet if it was rolled under conditions of simple slip, simply by electrolytically polishing the crystal and thus removing several mils of material from the surface.

Recovery, as measured by etch pit density, hardness, and Laue-back reflection micrographs, behaves in a similar manner. Complete recovery is most amenable under deformation by simple slip. But for multiple slip, the driving force for recrystallization is most favorable. Annealing temperatures in excess of 2000°C must be used to completely recover the original dislocation density and hardness; although some residual lattice bending is retained up to temperatures of 2800°C. With the times used, lower annealing temperatures near 1000°C, i.e., stress relieving causes no detectable changes in substructure although affecting ductility.

Another form of recovery is available which is more aptly described as recrystallization "in situ" i.e., completely softening prior to the formation of grain boundaries. This phenomenon occurs only under well defined rolling conditions and crystal orientations. On first analysis, it appears that the crystal orientation must be the same as the recrystallization texture found in the polycrystalline material, which is the (001) [110] for tungsten. Then the billet must be rolled such that there is very little change in crystal orientation. At most, lattice bending about one axis can be tolerated. Two crystal orientations satisfy this last prerequisite in tungsten. They are the (001) [110] and (110) [001]. Finally, the surface must be polished in order to remove all surface effects due to rolling.

In our program, experimental verification of this phenomenon has been obtained with the (001) [110] crystal reduced 50% and annealed at 1600°C for ½ hour. Similar results were nearly obtained with the (110) [001], which would negate the starting premise of only using the crystal with an orientation identical to the recrystallization texture of polycrystalline tungsten. Instead the second premise appears to be the controlling factor i.e., deformation must only occur by easy glide at the surfaces on conjugate slip systems which result in duplex slip in the center section of the thickness of the sheet. This last issue has yet to be substantiated but it seems highly reasonable on the basis of the over-all observations of this investigation. If true, the driving force for recrystallization can be used to obtain complete softening at temperatures near 1600°C instead of annealing temperatures approaching 2500°C, which is necessary for conventional recovery treatments.

5.3 ON THE EFFECT OF SUBSTRUCTURE ON TENSILE PROPERTIES

The success of this program depends on a rigorous understanding of the substructural changes accompanying deformation, recovery and recrystallization. The ultimate utilization of the product depends on an understanding of how the resulting substructure influences the mechanical properties. For the sake of discussion, the presentation will involve comments pertaining to the extreme case i.e., recrystallized tungsten and secondly to the intermediate case which shows the influence of warm work on the single crystal properties.

5.3.1 GRAIN BOUNDARIES

Single crystals are void of grain boundaries. By the process of recrystallization, grain boundaries are introduced into the crystal, thus producing polycrystalline tungsten. The influence of grain boundaries on the properties of tungsten can be evaluated by comparing the properties of monocrystalline and polycrystalline tungsten. Much data is already available for this evaluation;[12-14] but only the data from H. G. Sell[8] and coworkers will be cited to satisfy the need of this presentation. Their single crystals were produced by electron beam floating zone melting and the polycrystalline tungsten was recrystallized powder metallurgy tungsten processed from high purity (~ 99.98%) tungsten powders.

The absence of grain boundaries does not significantly alter the strength of tungsten. Over a temperature range of -150°F (-100°C) to 1475°F (800°C), the yield stress taken as the proportional limit is the same for both mono- and polycrystalline tungsten (Figure 26). The tensile strength of tungsten is reported as slightly higher for the polycrystalline tensile specimens. Below 300°F (150°C), no tensile data is available for the polycrystalline tungsten because of the brittle nature

of the material. Single crystal tungsten, on the other hand, can be tested in tension down to -150°F (-100°C) because of the lower ductile-to-brittle transition temperature. The latter results point to the most significant influence of grain boundaries -- eliminating room temperature ductility.

The effect of grain boundaries on low temperature ductility is more significant than the presence of impurity elements normally contained in the metal. The recrystallization of tungsten single crystals raises the bend transition temperature to about 575°F (300°C).[14] For both mono- and polycrystalline tungsten, as the impurity content increases, strength increases and ductility decreases.[15]

5.3.2 WARM WORK

Warm working can increase the strength of both mono- and polycrystalline tungsten. As discussed in this report, the degree to which work hardening is introduced into a single crystal must be refined to greater detail than for polycrystalline tungsten. The mode of slip must be considered for each single crystal orientation whereas gross effects are measured in rolled polycrystalline tungsten sheet. But for the sake of presenting a generalization, the influence of warm work on the strength and DBTT of mono- and polycrystalline tungsten is illustrated in Figure 27. Both increase in strength with an increase in percent warm work and correspondingly have a decrease in the recrystallization temperature.

The significant difference is the fact that increasing warm work lowers the DBTT for polycrystalline tungsten; but, it raises the DBTT of single crystal tungsten until they approach each other in the heavily worked condition.

Although work hardening decreases the DBTT of tungsten single crystal, their inherent ductility is such that the DBTT is initially as low as minus 150°F (-100°C). Therefore if the results of this program are utilized to fabricate single crystal sheet which possess a minimum amount of work hardening, sufficient room temperature ductility already exists. Furthermore, if annealing treatments can be used to remove a large portion of work hardening, the single crystal should have a recrystallization temperature approaching its melting point.

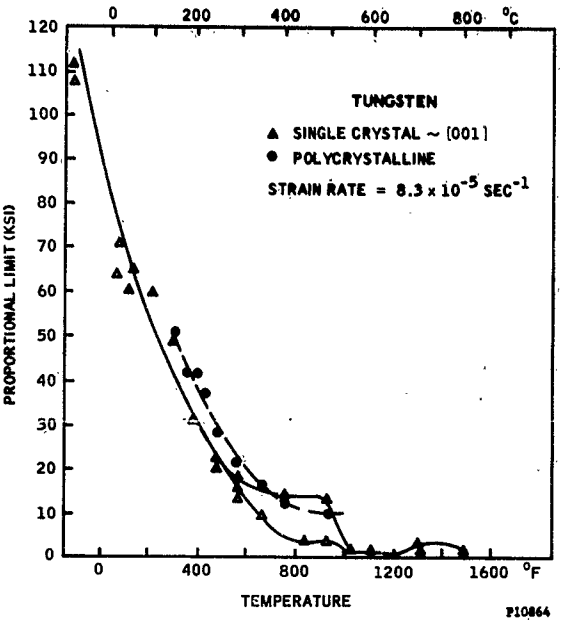

FIGURE 26. THE INFLUENCE OF GRAIN BOUNDARIES ON THE PROPORTIONAL LIMIT OF TUNGSTEN (After Sell, et al.[8])

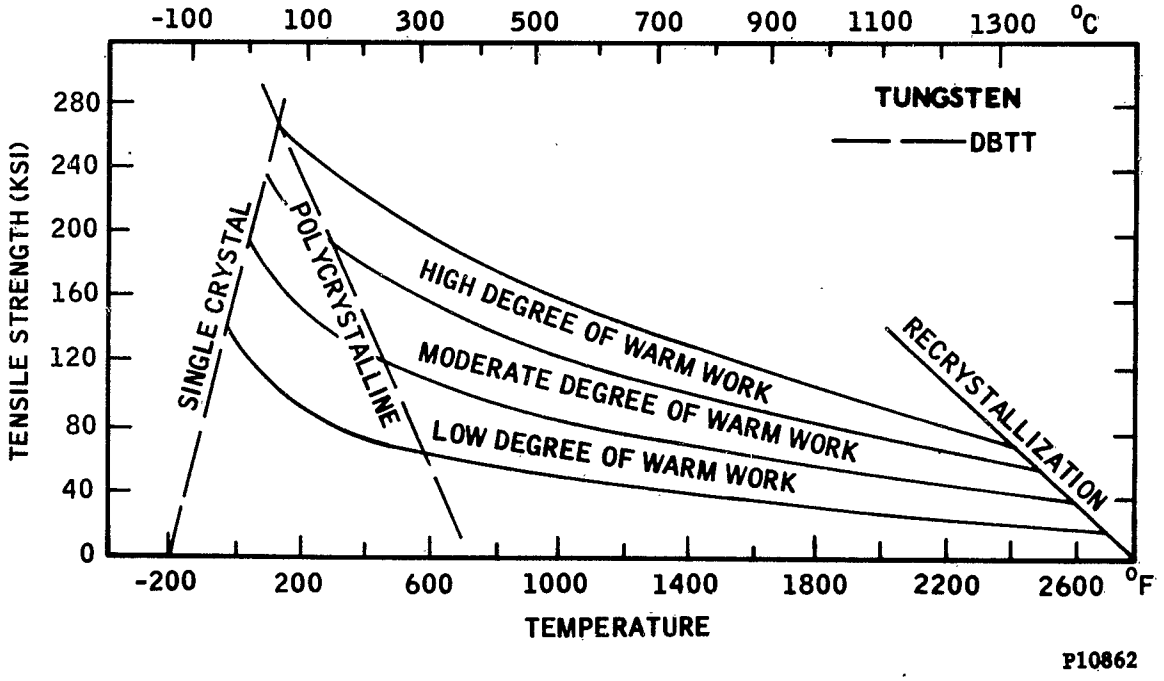

FIGURE 27. THE INFLUENCE OF WARM WORK ON THE STRENGTH AND DBTT OF MONO- AND POLYCRYSTALLINE TUNGSTEN (After Schmidt and Ogden[5])

SECTION 6

RECOMMENDATIONS FOR FUTURE WORK

(1) This year's program has advanced the single crystal sheet fabrication technology to the point that subsequent programs should be directed at manufacturing a piece of prototype hardware. The immediate effort should entail fabricating simple elements which can be used to evaluate various forming and joining procedures. With regard to forming, such operations as shearing, bending and possibly shear spinning should be considered. With regard to joining, preliminary results have already illustrated the tremendous advantages with electron beam welding. (See Figure 24). The details of the welding procedure should be established with due consideration to allowable tolerances, the amount and type of lattice distortion in the weld zone, weld schedules, tooling, and x-ray inspection techniques. Repair welding with the electron beam also appears to have advantages since this method of joining does not initiate recrystallization -- even in previously worked sections.

(2) The sensitivity of the mechanical properties and of the response to annealing to surface conditions has been illustrated in this report. More work should be done to clearly define this surface sensitivity. The amount of surface removal necessary for a reproducible response to annealing treatments should be determined. Tolerances should also be established by correlation with mechanical properties which are designed to evaluate notch sensitivity.

(3) Single crystal sheet fabrication technology can be further advanced by further research directed at understanding the influence of substitutional solid solution alloys on the deformation, recovery and

recrystallization processes of tungsten. These alloys would have immediate application because they provide high temperature strength.

(4) Finally, the technology should not be limited to tungsten. Already it is apparent that the electron beam welds in single crystals behave in a ductile fashion because of the absence of a recrystallized weld zone (See Figure 24). Embrittlement due to welding is a problem in other refractory metals such as molybdenum and tantalum. Hence, the restriction in the joining methods employed in the utilization of these metals could be removed by allowing greater flexibility in methods of fabrication.

SECTION 7

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